

CHAPTER 2

Criteria Pollutants-National Trends

<http://www.epa.gov/oar/aqtrnd97/chapter2.pdf>

This chapter presents national and regional trends for each of the pollutants for which the United States Environmental Protection Agency (EPA) has established National Air Quality Standards (NAAQS). NAAQS are in place for the following six criteria pollutants: carbon monoxide (CO), lead, nitrogen dioxide, ozone, particulate matter (PM), and sulfur dioxide (SO₂). Table 2-1 lists the NAAQS for each pollutant in terms of the level and averaging time of the standard used to evaluate compliance.

There are two types of standards: primary and secondary. Primary standards protect against adverse health effects, whereas secondary standards protect against welfare effects such as damage to crops, ecosystems, vegetation, buildings, and decreased visibility. There are primary standards for all of the criteria pollutants, and some pollutants (PM and SO₂) have primary standards for both long-term (annual average) and short-term (24 hours or less) averaging times. Short-term standards most directly protect people from any adverse health effects associated with peak short-term exposures to air pollution, while long-term standards can protect people from adverse health effects associated with short- and long-term exposures

Table 2-1. NAAQS in Effect in 1997

Pollutant	Primary (Health Related)		Secondary (Welfare Related)	
	Type of Average	Standard Level Concentration ^a	Type of Average	Standard Level Concentration
CO	8-hour ^b	9 ppm (10 mg/m ³)	No Secondary Standard	
	1-hour ^b	35 ppm (40 mg/m ³)	No Secondary Standard	
Pb	Maximum Quarterly Average	1.5 µg/m ³	Same as Primary Standard	
NO ₂	Annual Arithmetic Mean	0.053 ppm (100 µg/m ³)	Same as Primary Standard	
O ₃	1-hour ^c	0.12 ppm (235 µg/m ³)	Same as Primary Standard	
	8-hour ^d	0.08 ppm (157 µg/m ³)	Same as Primary Standard	
PM ₁₀	Annual Arithmetic Mean	50 µg/m ³	Same as Primary Standard	
PM _{2.5}	24-hour ^e	150 µg/m ³	Same as Primary Standard	
	Annual Arithmetic Mean ^f	15 µg/m ³	Same as Primary Standard	
	24-hour ^g	65 µg/m ³	Same as Primary Standard	
SO ₂	Annual Arithmetic Mean	0.03 ppm (80 µg/m ³)	3-hour ^b	0.50 ppm (1,300 µg/m ³)
	24-hour ^b	0.14 ppm (365 µg/m ³)		

^a Parenthetical value is an approximately equivalent concentration.

^b Not to be exceeded more than once per year.

^c Not to be exceeded more than once per year on average.

^d 3-year average of annual 4th highest concentration.

^e The pre-existing form is exceedance-based. The revised form is the 99th percentile.

^f Spatially averaged over designated monitors.

^g The form is the 98th percentile.

Source: 40 CFR Part 50.

to air pollution. Secondary standards have been established for each criteria pollutant except CO. Secondary standards are identical to the primary standard with the exception of SO₂.

On July 18, 1997, EPA issued the revised NAAQS for ozone and PM. The form of the revised ozone standard is the annual fourth highest 8-hour concentration averaged over 3 years. The level of the revised ozone NAAQS is 0.08 ppm. The form of the revised short-term PM₁₀ standard is the 99th percentile 24-hour average concentration averaged over 3 years. The level of the revised short-term PM₁₀ standard of 150 ug/m³ was retained. The form and level of the long-term PM₁₀ standard were retained. In addition, an indicator for PM_{2.5} was introduced to protect against particulate matter in the smaller end of the particle range. The form of the PM_{2.5} short-term standard is the 99th percentile 24-hour average concentration averaged over 3 years, and the associated level is 65 ug/m³. The form of the PM_{2.5} long-term standard is the annual arithmetic mean spatially averaged over designated monitors averaged over 3 years, and the associated level is 15 ug/m³. The revised standards are listed in Table 2-1 and are discussed in greater detail within the ozone and PM sections of this chapter.

The pre-existing standards for ozone are still in effect for areas that did not meet them prior to the NAAQS revisions. The pre-existing NAAQS for PM₁₀ are in effect until states take steps to remove them. Specifically, states must have a federally-approved State Implemen-

tation Plan (SIP) that shows how they can implement the revised PM NAAQS and all state adopted and implemented control measures. Both the pre-existing and the revised NAAQS are listed in Table 2-1 and trends associated with both are examined in this chapter.

Most of the trends information presented in this chapter is based on two types of data: ambient concentrations and emissions estimates. Ambient concentrations are measurements of pollutant concentrations in the ambient air from monitoring sites across the country. This year's report contains trends data accumulated from 1988 to 1997 on the criteria pollutants at 2,778 monitoring stations located throughout the United States. The trends presented here are derived from the composite average of these direct measurements. The averaging times and air quality statistics used in the trends calculations relate directly to the NAAQS.

The second type of data presented in this report reflects national emissions estimates. These are based largely on engineering calculations of the amounts and kinds of pollutants emitted by automobiles, factories, and other sources over a given period. In addition, some emissions estimates are based on measurements from continuous emissions monitors (CEMs) that have recently been installed at major electric utilities to measure actual emissions. This report incorporates data from CEMs collected between 1994 and 1997 for NO_x and SO₂ emissions at major electric utilities.

Changes in ambient concentrations do not always track changes in

emissions estimates. There are four known reasons for this. First, because most monitors are positioned in urban, population-oriented locales, air quality trends are more likely to track changes in urban emissions rather than changes in total national emissions. Urban emissions are generally dominated by mobile sources, while total emissions in rural areas may be dominated by large stationary sources such as power plants and smelters.

Second, emissions for some pollutants are calculated or measured in a different form than the primary air pollutant. For example, concentrations of ozone are caused by VOC emissions as well as NO_x emissions.

Third, the amount of some pollutants measured at monitoring locations depends on what chemical reactions, if any, occur in the atmosphere during the time it takes the pollutant to travel from its source to the monitoring station.

Finally, meteorological conditions often control the formation and buildup of pollutants in the ambient air. For example, peak ozone concentrations typically occur during hot, dry, stagnant summer-time conditions; CO is predominantly a cold weather problem; and the amount of rainfall can affect particulate matter levels and the frequency of forest fires.

For a more detailed discussion of the methodology used to compute the trends estimates in this chapter, please refer to Appendix B.

THE NATIONAL PERSPECTIVE: PAST, PRESENT, AND FUTURE

Improvements in the face of Economic Growth

National reductions in air quality concentrations and emissions continue to occur in the face of economic growth. Since 1970, total U.S. population increased 31 percent, vehicle miles traveled increased 127 percent, and the gross domestic product (GDP) increased 114 percent.^{1,2,3} During that same period, notable reductions in air quality concentrations and emissions took place. Aggregate criteria pollutant emissions decreased 31 percent. When examined individually, emissions for all criteria pollutants except NO_x decreased between 1970 and 1997, the greatest improvement being a 98-percent decrease in lead emissions. Though air quality trends are not available back to 1970, in most cases they are available for the past 20 years. Reductions in air quality concentrations between 1978 and 1997 are impressive with CO, lead, and SO₂ decreasing by more than half.⁴ These air quality improvements are a direct result of EPA working with states, industry, and other partners to effectively establish and implement clean air laws and regulations.

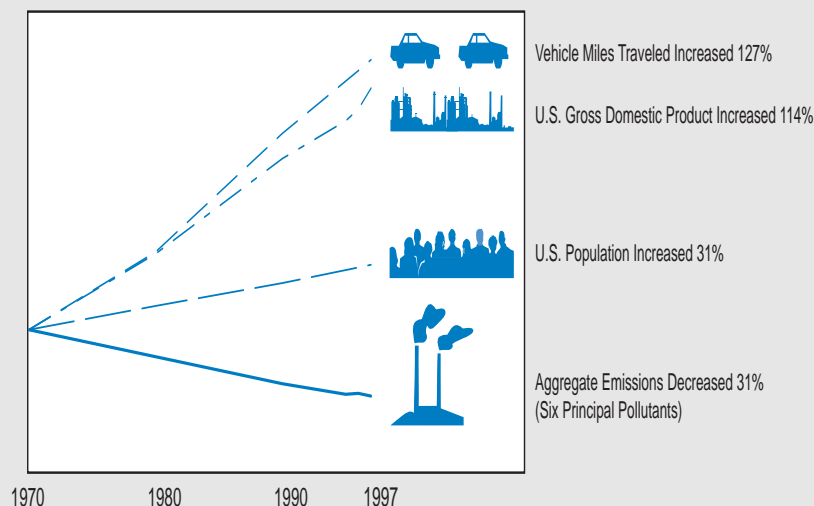
The Need for Continued Progress

While progress has been made, it is important not to lose sight of the air pollution problems that still remain. Though air quality trends are improving nationally, there are still areas, both urban and rural, with concentrations above the level of the national standard and even areas with worsening trends (in addition to Chapter 2, see Chapter 3 for more details on urban areas and Chapter 6 for more details on rural areas). Based upon monitoring data submitted to EPA's Aerometric Information Retrieval System (AIRS) data base, approximately 107 million people in the United States reside in counties that did not meet the air quality standard for at least one of the NAAQS pollutants for the single year 1997.^{5,6}

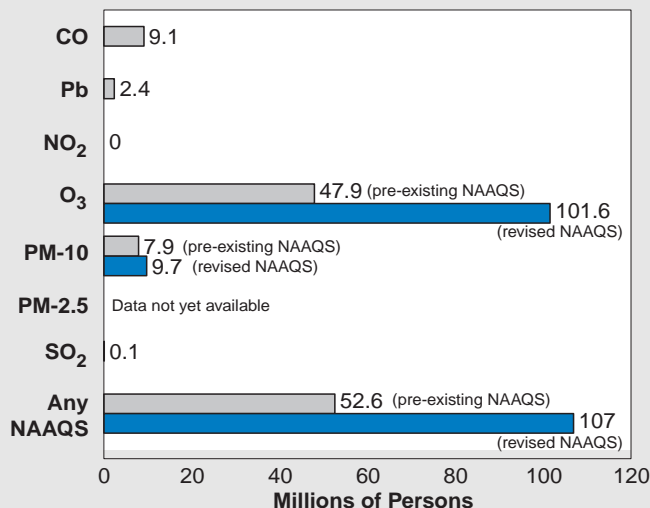
Long-term Percent Change in National Air Quality Concentration and Emissions

	Air Quality Concentration %Change 1978-1997	Emissions % Change 1970-1997
CO	-60%	-32%
Pb	-97%	-98%
NO ₂	-25%	+11%
O ₃	-30% (1hr)	-37%
PM ₁₀	Data Not Available	-75%
SO ₂	-55%	-35%

*Includes only directly emitted particles. Secondary PM formed from SO_x, NO_x, and other gases comprise a significant fraction of ambient PM.



Total U.S. Population, vehicle miles traveled, U.S. gross domestic product, and aggregate emissions, 1970-1997.



Number of people living in counties with air quality concentrations above the level of the NAAQS in 1997.

1. Statistical Abstract of the United States, 1996, U.S. Department of Commerce, U.S. Bureau of Census.
2. E.H. Pechan & Associates, Springfield, VA, October 1998.
3. The Bureau of Economic Analysis, Department of Commerce, website at <http://www.bea.doc.gov/bea/>.
4. Because of evolving monitoring networks, these long-term changes in air quality concentrations are not as certain as the more recent 10-year assessment.
5. The population estimates are based upon only a single year of data, 1997, and only consider counties with monitoring data for each pollutant. They are intended to provide a relative measure of the extent of the problem for each pollutant in 1997. An individual living in a county that had a measured concentration above the level the NAAQS may not actually be exposed to unhealthy air.
6. The number of people living in formally designated nonattainment areas as of September 1998 was approximately 113 million. These population estimates differ because formal nonattainment designations are based on multiple years of data rather than a single year and generally do not follow county boundaries. For a pollutant such as ozone, nonattainment areas typically compose the entire metropolitan area, which may include additional counties that do not contain monitors. Also, designations have not yet been made for the revised ozone and PM NAAQS. Therefore, the nonattainment area population does not reflect the revised NAAQS.

CARBON MONOXIDE

• Air Quality Concentrations

1988–97	38% decrease
1996–97	7% decrease

• Emissions

1988–97	25% decrease
1996–97	3% decrease

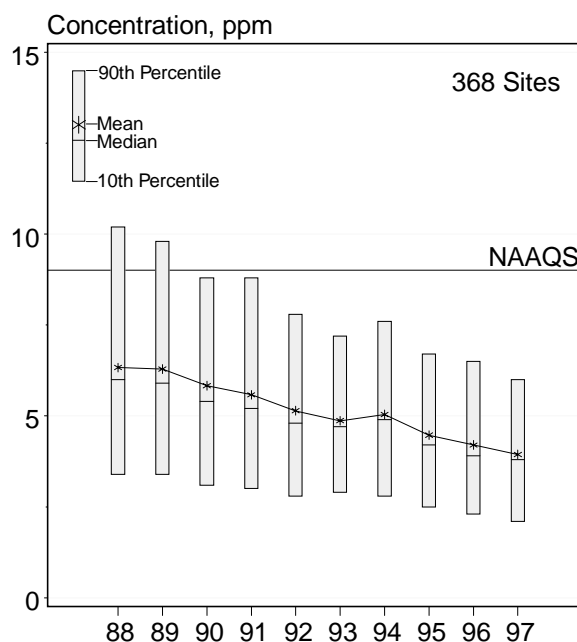
Nature and Sources

CO is a colorless, odorless, and at much higher levels, a poisonous gas formed when carbon in fuels is not burned completely. It is a product of motor vehicle exhaust, which contributes about 60 percent of all CO emissions nationwide. High concentrations of CO generally occur in areas with heavy traffic congestion. In cities, as much as 95 percent of all CO emissions may emanate from automobile exhaust. Other sources of CO emissions include industrial processes, non-transportation fuel combustion, and natural sources such as wildfires. Woodstoves, cooking, cigarette smoke, and space heating are sources of CO in indoor environments. Peak CO concentrations typically occur during the colder months of the year when CO automotive emissions are greater and nighttime inversion conditions are more frequent.

Health Effects

Carbon monoxide enters the bloodstream through the lungs and reduces oxygen delivery to the body's organs and tissues. The health threat from lower levels of CO is most serious for those who suffer from cardiovascular disease, such as angina pectoris. At much higher levels of exposure, CO can be poisonous, and healthy individuals may also be affected. Visual impairment, reduced work capacity,

Figure 2-1. Trend in second maximum non-overlapping 8-hour average CO concentrations, 1988-1997.



reduced manual dexterity, poor learning ability, and difficulty in performing complex tasks are all associated with exposure to elevated CO levels.

Primary Standards

There are two primary NAAQS for ambient CO, a 1-hour average of 35 ppm and an 8-hour average of 9 ppm. These concentrations are not to be exceeded more than once per year. There currently are no secondary standards for CO.

National 10-Year Trends

The use of a moving 10-year trends window maximizes the number of sites that meet the minimum data completeness requirement of 8 years of ambient monitoring data. The 10-year trend in ambient CO concentrations is graphically shown in Figure 2-1. Air quality improvement is clear given the decreases shown in

CO concentrations for “peak” sites, “typical” sites, and “clean” sites (i.e., the 90th percentile, the composite mean, and the 10th percentiles.) Nationally, CO concentrations decreased 38 percent during the past 10 years as measured by the composite average of the annual second highest 8-hour concentration. Between 1996 and 1997, national composite average CO concentrations decreased 7 percent. Nationally, the 1997 composite average 8-hour ambient CO concentration is the lowest level recorded during the past 10 years.

Figure 2-2 presents diurnal patterns in hourly CO concentrations averaged across all sites for the 10-year period, 1987–96. The figure shows that reductions in CO concentrations are not limited to the peak hours, but are found at all hours throughout the day.¹

Reductions in ambient CO concentrations also occurred across all

monitoring environments—urban, suburban, and rural sites. Figure 2-3 shows that urban monitoring sites record higher CO concentrations on average, than suburban sites, with the lowest levels found at 12 rural CO sites. During the past 10 years, composite mean CO 8-hour concentrations decreased 39 percent at 208 urban sites, 35 percent at 145 suburban locations, and 46 percent at 12 rural monitoring sites.

Emissions Trends

Figure 2-4 shows that national total CO emissions have decreased 25 percent since 1988. Emissions from all transportation sources have decreased 22 percent during the past 10 years, despite a 25 percent increase in vehicle miles traveled (VMT). Because the urban CO monitoring sites are primarily mobile-source oriented, the 38 percent reduction in CO concentrations more closely tracks the estimated 29 percent reduction in emissions from highway vehicles. Figure 2-6 shows that the transportation category, composed of on-road and off-road sources, accounts for 77 percent of the nation's total CO emissions in 1997. Total CO emissions decreased 2 percent since 1996, with CO emissions from highway vehicles recording a 6-percent decline since last year, while VMT increased by 2 percent since 1996.

Table 2-2 lists some of the major milestones in the control of emissions from automobiles starting with the Clean Air Act (CAA) of 1970. At the national level, these measures include establishing national standards for tailpipe emissions, new vehicle technologies, and clean fuels programs. These measures have led to reductions in emissions of CO as well as

Figure 2-2. Diurnal plot of mean winter (December-February) hourly CO concentrations, 1987-1996.

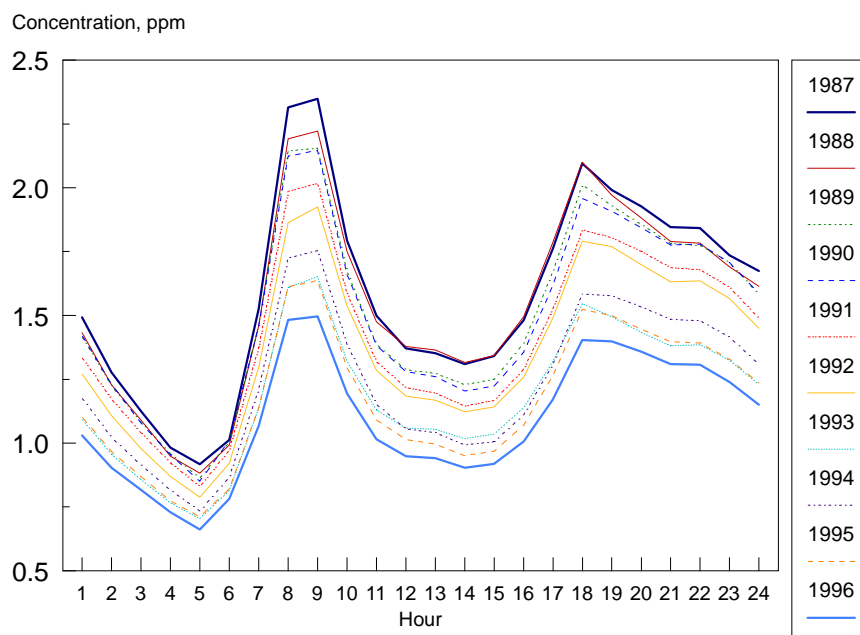


Figure 2-3. Trend in second maximum non-overlapping 8-hour average CO concentrations by type of location, 1988-1997.

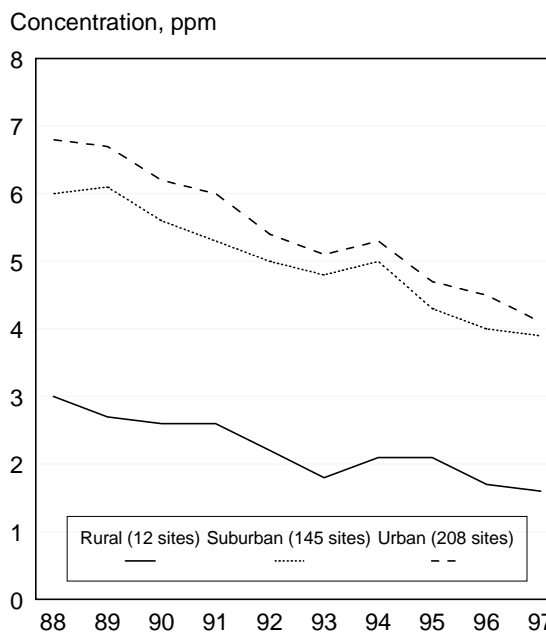
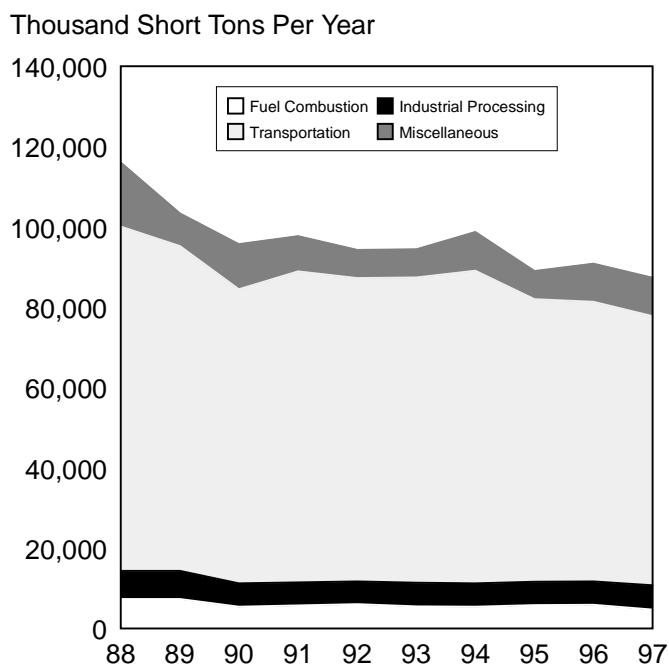
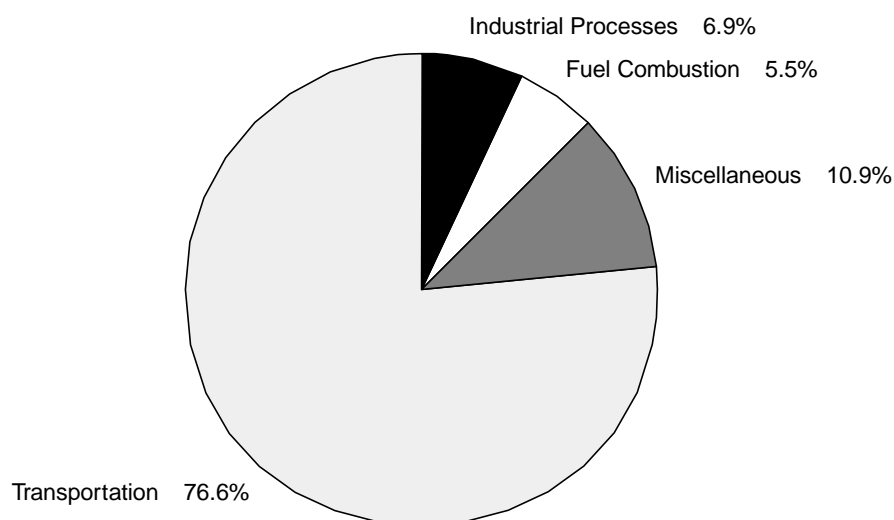


Figure 2-4. Trend in national total CO emissions, 1988-1997.**Figure 2-5.** CO emissions by source category, 1997.

other pollutants. State and local emissions reduction measures include inspection and maintenance (I/M) programs and transportation management programs.

Table 2-2. Milestones in Auto Emissions Control

1970	New Clean Air Act sets auto emissions standards.
1971	Charcoal canisters appear to meet evaporative standards.
1972	EGR valves appear to meet NO _x standards.
1974	Fuel economy standards are set.
1975	The first catalytic converters appear for hydrocarbon, CO. Unleaded gas appears for use in catalyst equipped cars.
1981	3-way catalysts with on-board computers and O ₂ sensors appear.
1983	I/M programs are established in 64 cities.
1989	Fuel volatility limits are set for RVP.
1990	CAAA set new tailpipe standards.
1992	Oxyfuel introduced in cities with high CO levels.
1993	Limits set on sulfur content of diesel fuel.
1994	Phase-in begins of new vehicle standards and technologies.

In the area of clean fuels, the 1990 Clean Air Act Amendments (CAAA) require oxygenated gasoline programs in several regions during the winter months. Under the program regulations, a minimum oxygen content (2.7 percent by weight) is required in gasoline to ensure more complete fuel combustion.^{2,3} Of the 36 nonattainment areas that initially implemented the program in 1992, 25

areas continue to use oxygenated fuels. An analysis of the oxygenated fuels program in several cities with winter oxygenated gasoline programs showed reductions in ambient CO concentrations of about 10 percent.⁴ Other studies estimated that the oxyfuel effect was an average total reduction in ambient CO concentrations of 7 to 14 percent overall for the eight winter seasons from 1986 through 1994.^{5,6}

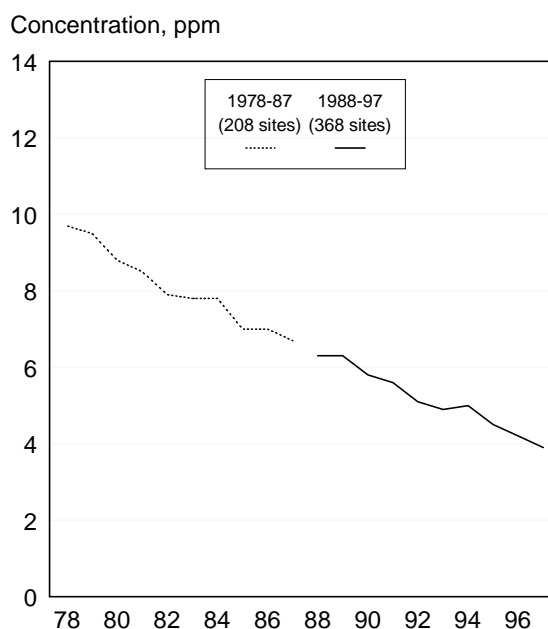
National 20-Year Trends

Because of the annual loss and replacement of ambient monitoring sites, too few sites are able to meet a 20-year data completeness criteria. Thus, long-term trends are assessed by piecing together two separate 10-year trends data bases. Because the number of monitoring sites nationwide can vary greatly over a 20-year period, two 10-year periods are used to capture a common set of sites for each period. Although there are differences in the mix of trend sites for the two periods (208 vs. 367 sites), Figure 2-6 shows a consistent decline in CO concentrations during the past 20 years. Nationally, the 1997 composite average ambient concentration is 60 percent lower than 1978 and the lowest level recorded during the past 20 years of monitoring.

Regional Trends

The map in Figure 2-7 shows the regional trends in ambient CO concentrations during the past 10 years, 1988–1997. All 10 EPA Regions recorded 10-year declines in CO levels as measured by the regional composite mean concentrations. The largest concentration reductions are in the Northcentral, Rocky Mountain and

Figure 2-6. Long-term trend in second maximum non-overlapping 8-hour average CO concentrations, 1978–1997.



Northwest states. Smaller reductions can be seen in the West, South and Midwest regions. Only the Southeast (Region 4) saw an upturn between 1996 and 1997 (an increase of 3 percent).

1997 Air Quality Status

The map in Figure 2-8 shows the variations in CO concentrations across the country in 1997. The air quality indicator is the highest annual second maximum 8-hour concentration measured in each county. The bar chart to the left of the map displays the number of people living in counties within each concentration range. The colors on the map and bar chart correspond to the colors of the concentration ranges displayed in the map legend. Only 6 of the 537 monitoring sites reporting ambient CO data to AIRS failed to meet the CO NAAQS in 1997. These six sites were located in three counties with a total

population of 9 million people—Los Angeles County, CA; Fairbanks, AK; and Imperial County, CA (Calexico, CA). The site in this latter area is located ¼ mile north of the border crossing with Mexicali, Mexico. This is an improvement over the 1996 totals of seven counties with a total population of 13 million people.

Data Sources

The CO ambient trends plotting points and emissions totals by source category are listed in Tables A-1 and A-2. The plotting points for the 20-year trend charts are listed in Table A-9. The 1997 county maximum second-highest non-overlapping 8-hour CO concentrations are listed in Table A-11.

Figure 2-7. Trend in CO second maximum 8-hour concentrations by EPA Region, 1988-97.

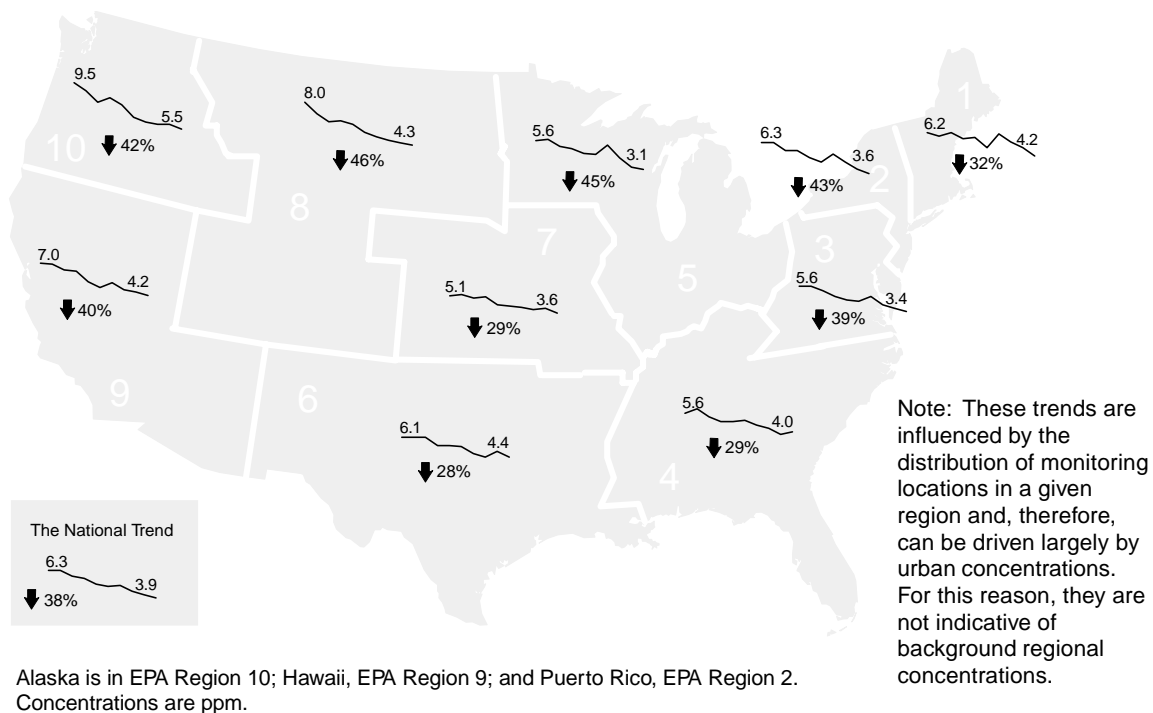
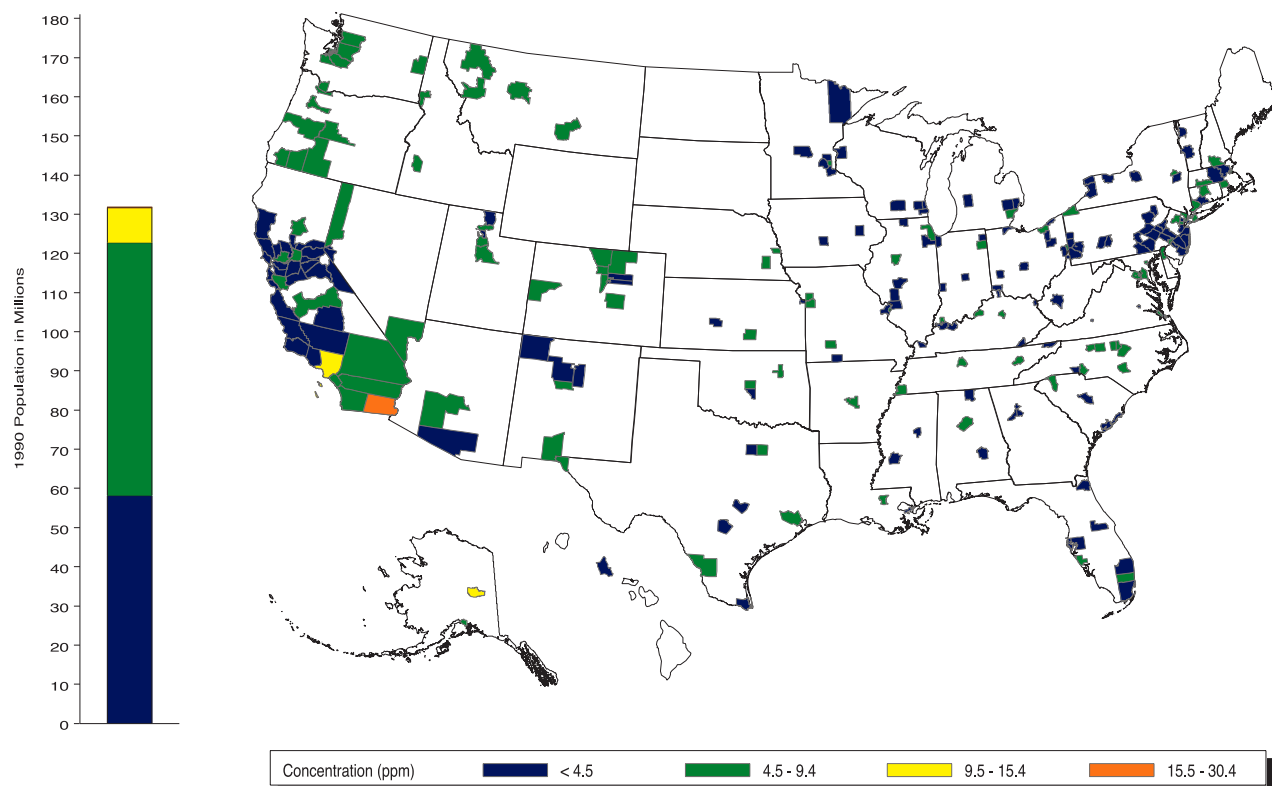


Figure 2-8. Highest second maximum non-overlapping 8-hour average CO concentration by county, 1997.



LEAD

• Air Quality Concentrations

1988–97	67% decrease
1996–97	no change

• Emissions

1988–97	44% decrease
1996–97	no change

Nature and Sources

Twenty years ago, automotive sources were the major contributor of lead emissions to the atmosphere. As a result of EPA's regulatory efforts to remove lead from gasoline, the contribution from the transportation sector has dramatically declined. Today, metals processing is the major source of lead emissions to the atmosphere. The highest ambient air concentrations of lead are found in the vicinity of ferrous and nonferrous smelters, battery manufacturers, and other stationary sources of lead emissions.

Health and Environmental Effects

Exposure to lead occurs mainly through inhalation and ingestion of lead in food, water, soil, or dust. It accumulates in the blood, bones, and soft tissues. Lead also can adversely affect the kidneys, liver, nervous system, and other organs. Excessive exposure to lead may cause neurological impairments such as seizures, mental retardation, and/or behavioral disorders. Even at low doses lead exposure is associated with changes in fundamental enzymatic, energy transfer, and homeostatic mechanisms in the body. At low doses fetuses and children may suffer from central nervous system damage. Recent studies show that lead may be a factor in high blood pressure and subsequent heart disease. Recent

studies also indicate that neuro-behavioral changes may result from lead exposure during the child's first years of life.

Airborne lead also can have adverse impacts on the environment. Wild and domestic grazing animals may ingest lead that has deposited on plant or soil surfaces or that has been absorbed by plants through leaves or roots. Animals, however, do not appear to be more susceptible or more sensitive to adverse effects from lead than humans. For this reason, the secondary standard for lead is identical to the primary standard. At relatively low concentrations (2–10 $\mu\text{g}/\text{m}^3$), lead can inhibit plant growth and lead to a shift to more tolerant plant species near roadsides and stationary source emissions.

In spite of the fact that the majority of soil lead becomes bound so that it is insoluble, immobile, and biologically unavailable, elevated soil lead concentrations have been observed to cause shifts in the microbial community (fungi and bacteria), reduced numbers of invertebrates, reduced decomposition and nitrification rates, and alterations in other soil parameters. Because lead remains in the soil, soil concentrations continue to build over time even when deposition rates are low. Thus, another concern is that acid precipitation may be increasing the mobility and bioavailability of soil lead in some places.

Lead enters water systems mainly through urban runoff and sewage and industrial effluents. Most of this lead is readily complexed and bound in the sediment. However, water lead concentrations can reach levels that are associated with increased mortality and impaired reproduction in aquatic invertebrates

and blood and neurological changes in fish. Given the above effects, there continue to be implications for the long-term impact of lead on ecosystem function and stability. [See also Chapter 5: Air Toxics and the December 1990 *OAQPS Staff Paper* (EPA-450/2-89-022)].

Primary and Secondary Standards

The primary and secondary NAAQS for lead is a quarterly average concentration not to exceed 1.5 $\mu\text{g}/\text{m}^3$.

National 10-Year Trends

The statistic used to track ambient lead air quality is the maximum quarterly mean concentration of each year. A total of 195 ambient lead monitors met the trends data completeness criteria. Point source-oriented monitoring data were excluded from all ambient trends analyses presented in this section so as not to mask the underlying urban trends. Figure 2-9 indicates that between 1988 and 1997, maximum quarterly average lead concentrations decreased 67 percent at population-oriented monitors. The decline was fairly similar at rural, suburban, and urban locations as seen in Figure 2-10. Between 1996 and 1997, national average lead concentrations (approaching the minimum detectable level) remained unchanged.

Emissions Trends

Figure 2-11 shows that total lead emissions decreased 44 percent between 1988 and 1997. The large ambient and emissions reductions are a waning result of the phase-out of leaded gasoline. Table A-3, which lists lead emissions by major source category, shows that on-road vehicles accounted for 82 percent of the 10-

Figure 2-9. Trend in maximum quarterly average Pb concentrations (excluding source-oriented sites), 1988-1997.

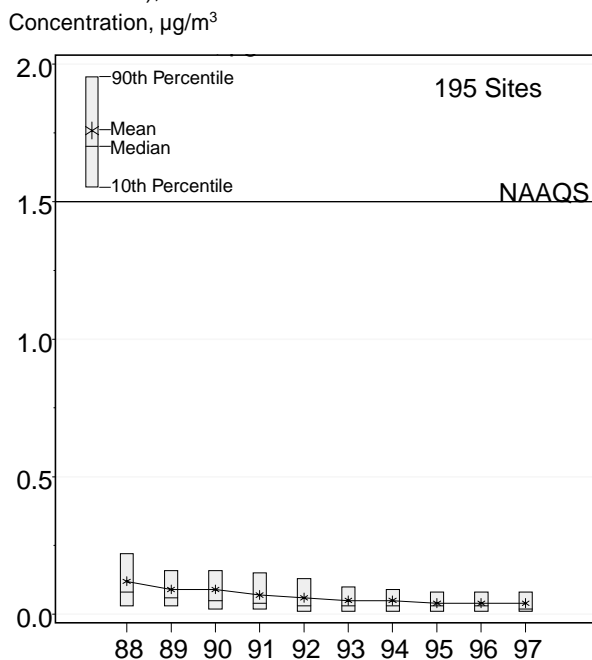
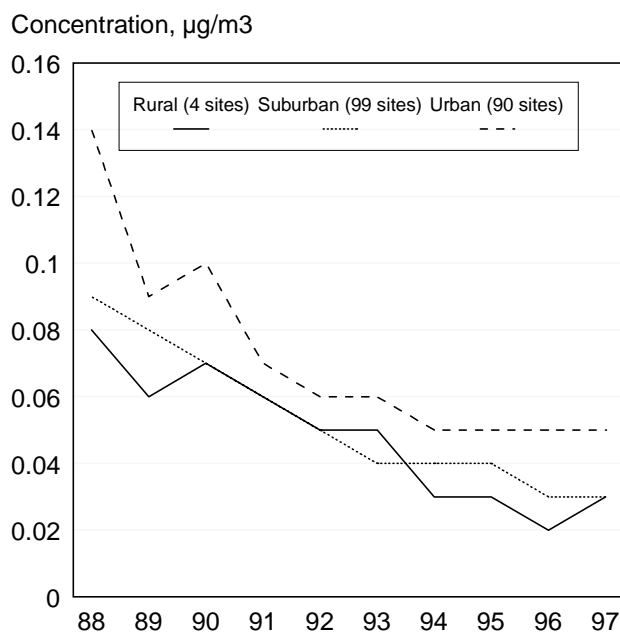


Figure 2-10. Pb maximum quarterly mean concentration trends by location (excluding source-oriented sites), 1988-1997.



year emissions decline. Between 1996 and 1997, lead emissions did not change substantially. Figure 2-12 shows that industrial processes were the major source of lead emissions in 1997, accounting for 74 percent of the total. The transportation sector (on-road and non-road sources) now accounts for only 13 percent of total 1997 lead emissions; on-road vehicles account for only one-half of a percent.

National 20-Year Trends

The effect of the conversion to unleaded gasoline usage on ambient lead concentrations is most impressive when viewed over a longer period, such as illustrated in Figure 2-13. Between 1978 and 1997, ambient concentrations of lead declined 97 percent. This large decline tracks well with the overall emissions trend, which shows a decline of 98 percent between 1975 and 1997.

Regional Trends

Figure 2-14 segregates the ambient trend analysis by EPA region. Although all regions showed large concentration reductions between 1988 and 1997, there were some intermittent upturns. Many of the latter year upturns and dips can be attributed to the inherent variability associated with data reported near the instrument's lower limit of detection.

1997 Air Quality Status

The large reductions in long-term lead emissions from transportation sources has changed the nature of the ambient lead problem in the United States. Because industrial processes are now responsible for all violations of the lead standard, the lead monitoring strategy now focuses on these emissions point sources. The map in

Figure 2-15 shows the lead monitors oriented in the vicinity of major sources of lead emissions. In 1997, four lead point sources had one or more source-oriented monitors that violated the NAAQS. These four sources are ranked in Figure 2-15 according to the site with greatest maximum quarterly mean. Various enforcement and regulatory actions are being actively pursued by EPA and the states for these sources.

The map in Figure 2-16 shows the highest quarterly mean lead concentration by county in 1997. Four counties, with a total population of 2.4 million and containing the point sources identified in Figure 2-15, did not meet the lead NAAQS in 1997.

Monitoring Status

Because of the shift in ambient air monitoring focus from mobile source emissions to stationary point sources of lead air pollution, EPA has taken action to revise the lead air monitoring regulations. This action is being taken at the direct request of numerous State and local agencies whose on-road mobile source-oriented lead monitors have been reporting peak lead air pollution values that are many times less than the quarterly lead NAAQS of $1.5 \mu\text{g}/\text{m}^3$ for a number of consecutive years. EPA published a direct final rule for ambient air quality surveillance for lead on November 5, 1997 in the *Federal Register*. However, due to adverse comments received, the rule was withdrawn on December 23, 1997. It is anticipated that the final rule will be published in late December 1998. The previous (current) regulation requires that each urbanized area with a population of 500,000 or more operate at least two lead National Air

Figure 2-11. National total Pb emissions trend, 1988-1997.

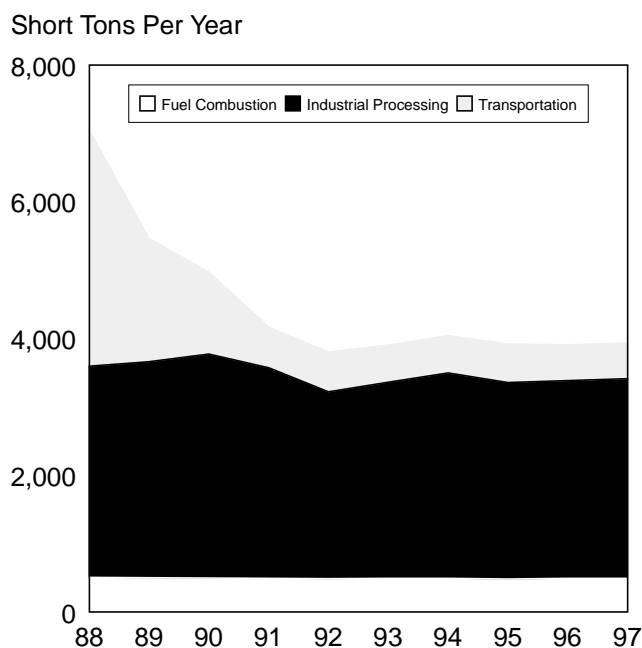


Figure 2-12. Pb emissions by source category, 1997.

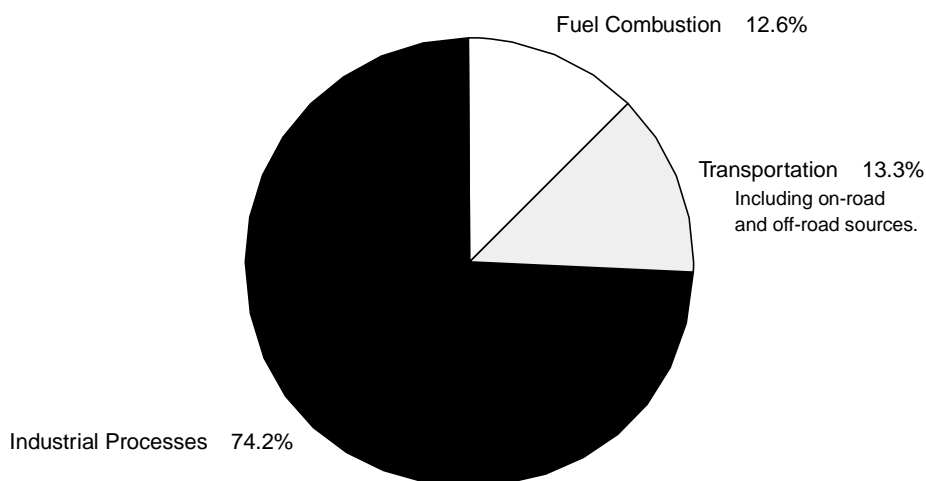
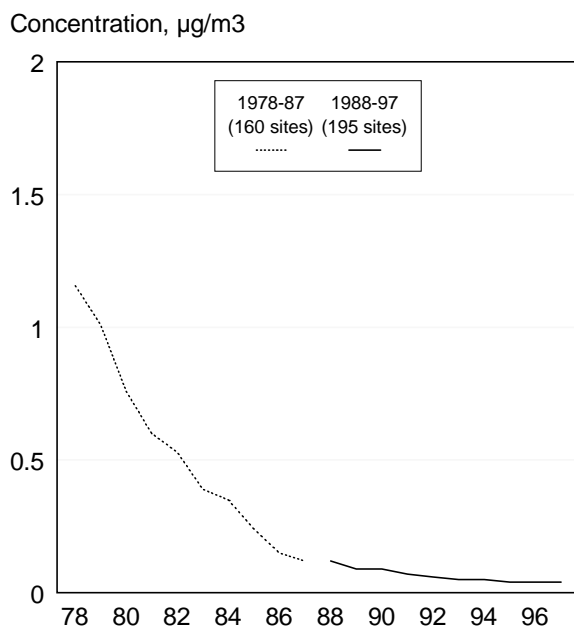


Figure 2-13. Long-term ambient Pb trend, 1977-1997.



Monitoring Stations (NAMS). The new lead monitoring rule maintains a minimum number of traditional types of lead monitoring sites in the largest metropolitan areas, and it refocuses available monitoring resources into areas with industrial sources.

Figure 2-14. Trend in Pb maximum quarterly mean concentrations by EPA Region, 1988-1997.

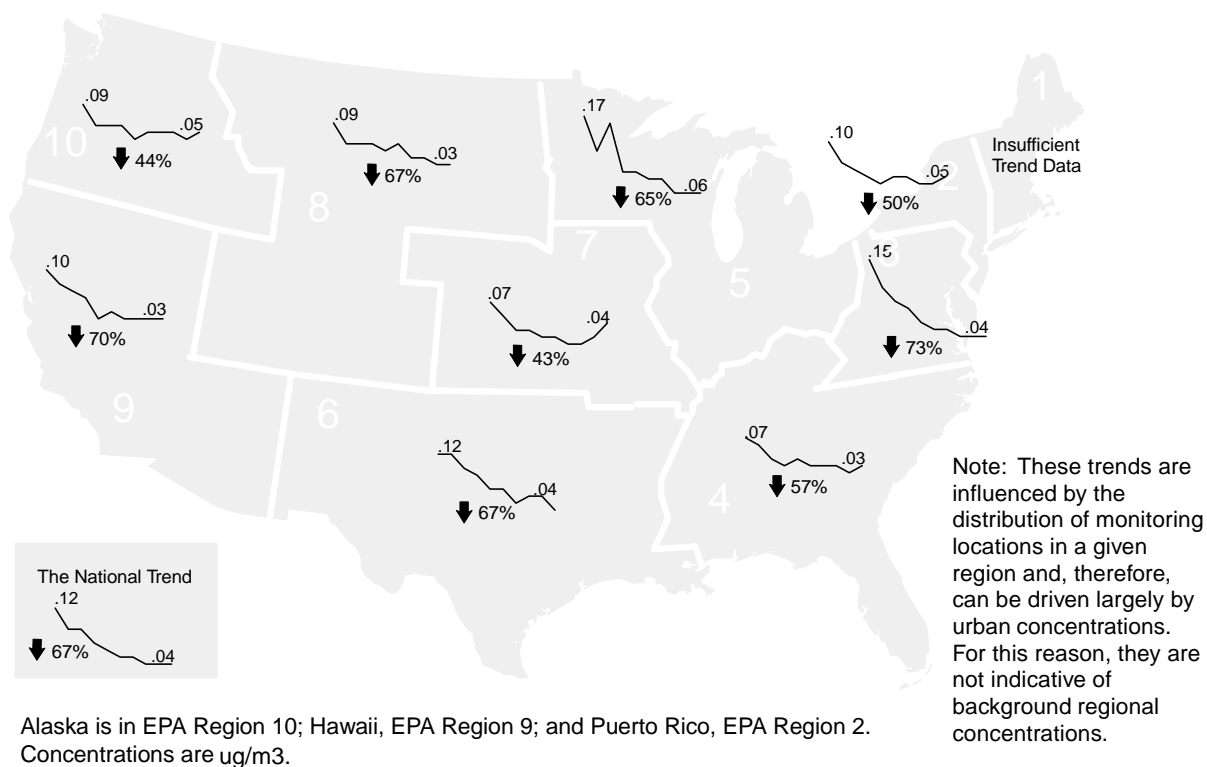
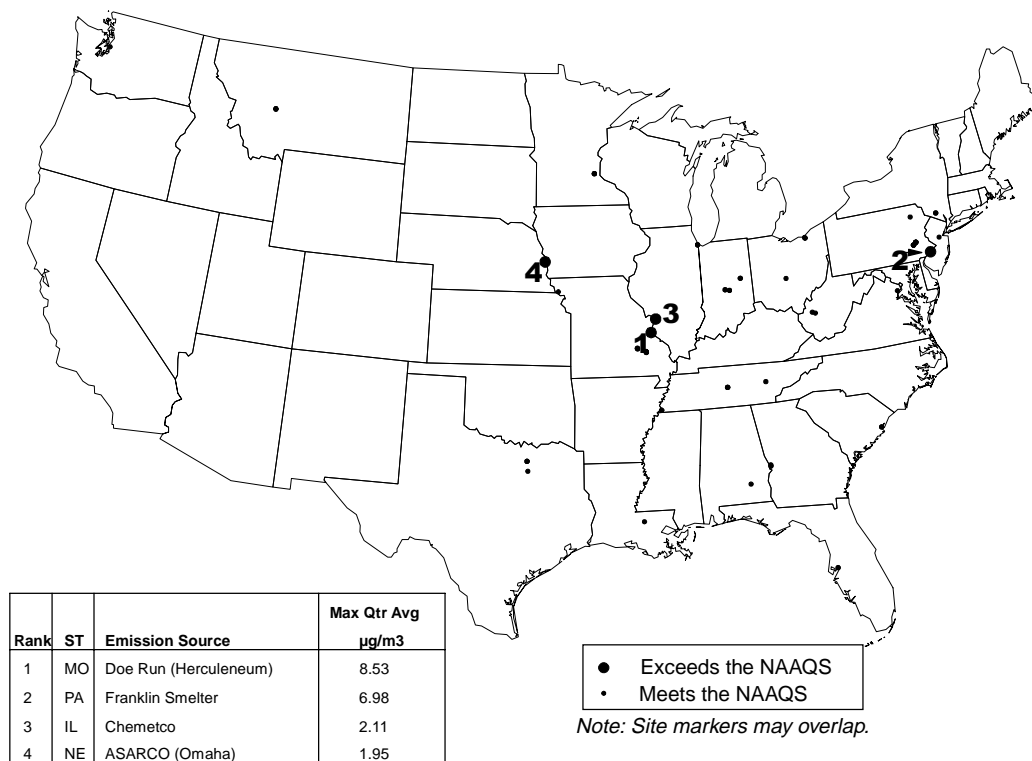
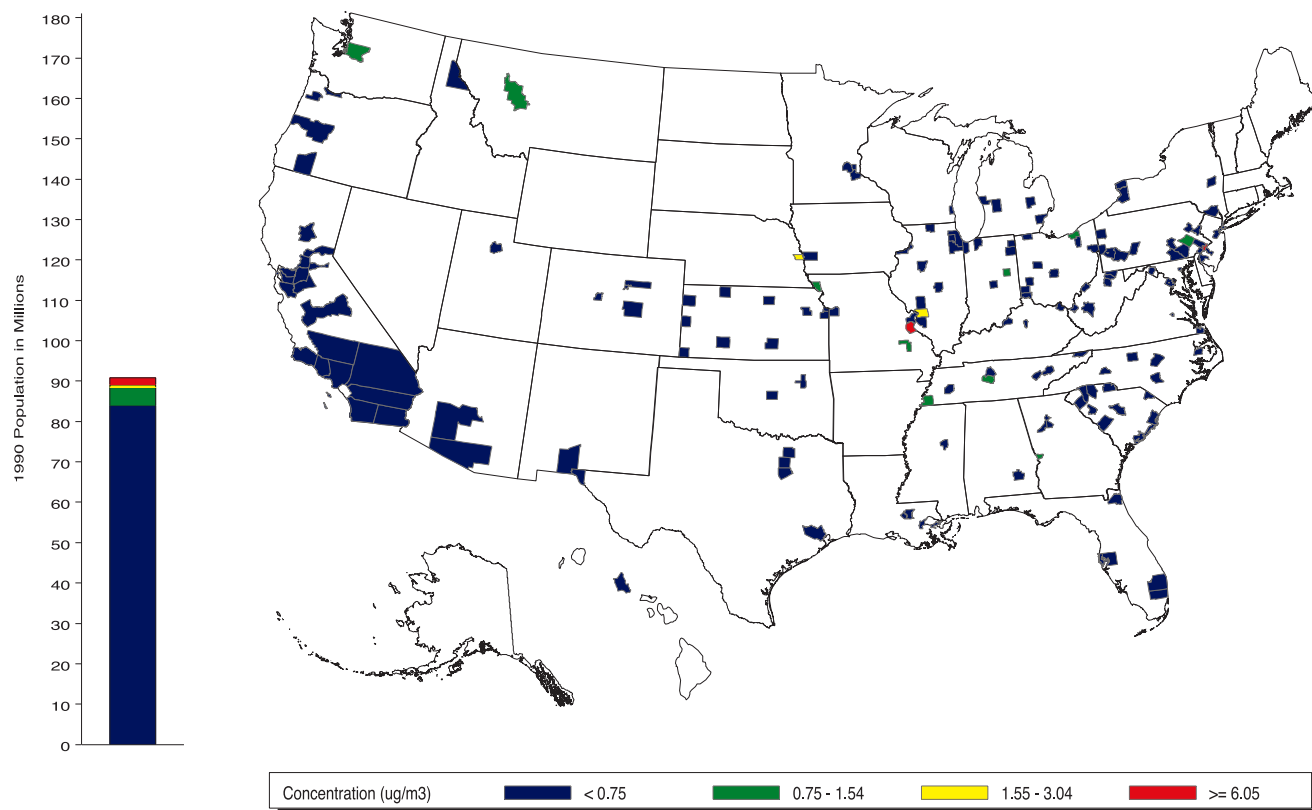


Figure 2-15. Pb maximum quarterly concentration in the vicinity of Pb point sources, 1997.**Figure 2-16.** Highest Pb maximum quarterly mean by county, 1997.

NITROGEN DIOXIDE

• Air Quality Concentrations

1988–97	14% decrease
1996–97	no change

• Emissions

1988–97	1% decrease
1996–97	1% increase

Nature and Sources

Nitrogen dioxide (NO₂) is a reddish brown, highly reactive gas that is formed in the ambient air through the oxidation of nitric oxide (NO). Nitrogen oxides (NO_x), the term used to describe the sum of NO, NO₂ and other oxides of nitrogen, play a major role in the formation of ozone in the atmosphere through a complex series of reactions with volatile organic compounds. A variety of NO_x compounds and their transformation products occur both naturally and as a result of human activities. Anthropogenic (i.e., man-made) emissions of NO_x account for a large majority of all nitrogen inputs to the environment. The major sources of anthropogenic NO_x emissions are high-temperature combustion processes, such as those occurring in automobiles and power plants. Most (about 95 percent) of NO_x from combustion sources is emitted as NO; the remainder is largely NO₂. Because NO is readily converted to NO₂ in the environment, the emissions estimates reported here assume nitrogen oxides are in the NO₂ form. Natural sources of NO_x are lightning, biological and abiological processes in soil, and stratospheric intrusion. Ammonia and other nitrogen compounds produced naturally are important in the cycling of nitrogen through the ecosystem. Home heaters and gas stoves also produce substantial amounts of NO₂ in indoor settings.

Health and Environmental Effects

NO₂ is the only nitrogen oxide sufficiently widespread and commonly found in ambient air at high enough concentrations to be a matter of public health concern. The health effects of most concern associated with short-term exposures (e.g., less than 3 hours) to NO₂ at or near the ambient NO₂ concentrations seen in the United States include changes in airway responsiveness and pulmonary function in individuals with pre-existing respiratory illnesses and increases in respiratory illnesses in children (5–12 years old).^{7,8}

Evidence suggests that long-term exposures to NO₂ may lead to increased susceptibility to respiratory infection and may cause alterations in the lung. Atmospheric transformation of NO_x can lead to the formation of ozone and nitrogen-bearing particles (e.g., nitrates and nitric acid). As discussed in the ozone and PM sections of this report, exposure to both PM and ozone is associated with adverse health effects.

Nitrogen oxides contribute to a wide range of effects on public welfare and the environment including potential changes in the composition and competition of some species of vegetation in wetland and terrestrial systems, and visibility impairment. However, the role nitrogen deposition plays in the acidification of freshwater bodies and the eutrophication of estuarine and coastal waters (e.g., Chesapeake Bay) is the deposition-related issue of most concern in the United States. Adverse environmental effects include the loss or shift in number and type of species, and explosive algae growth leading to a depletion of oxygen in the water, and/or an increase in levels of toxins

harmful to fish and other aquatic life. Nitrogen oxides are an important precursor both to ozone and to acidic deposition (see sections on ozone and sulfur dioxide trends.) NO_x emissions also can have a significant impact on particulate matter concentrations, most notably in some western urban areas.

Primary and Secondary Standards

The level for both the primary and secondary national ambient air quality standards (NAAQS) for NO₂ is 0.053 ppm annual arithmetic average, not to be exceeded.

National 10-Year Trends

The statistic used to track ambient NO₂ air quality trends is the annual mean NO₂ concentration. A total of 224 ambient NO₂ monitoring sites met the trends data completeness criteria. The national trend in annual mean NO₂ concentrations is shown graphically in Figure 2-17 for the 10-year period, 1988–1997. Based on measurements at 224 monitoring sites located in cities throughout the country, the 1997 national composite mean NO₂ concentration is 14 percent lower than the composite mean recorded in 1988, and is unchanged from the 1996 level. Figure 2-17 shows that sites recording the highest annual mean NO₂ concentrations (the 90th percentile) have recorded the largest reductions. Except for 1994, annual mean NO₂ concentrations have decreased yearly since 1989. Figure 2-18 shows how the trends in annual mean NO₂ concentrations vary among urban, suburban and rural monitoring locations. As Figure 2-18 illustrates, the highest annual mean NO₂ concentrations are typically found in urban areas, with significantly lower annual

mean concentrations recorded at rural sites. Trends in annual mean NO_2 concentrations are similar at both urban and suburban sites. The 1997 composite mean at 80 urban sites is 19 percent lower than the 1988 level, compared to 17 percent lower at 96 suburban sites. At 46 rural sites, the composite mean NO_2 concentration decreased in 1990 and remained constant for the next 4 years. The 1997 composite mean NO_2 concentration at these rural sites is 22 percent lower than the 1988 composite mean level. (See Figure B-3 in Appendix B for a map of the NO_2 monitoring site locations.)

Atmospheric concentrations of NO_2 are determined by indirect photomultiplier measurement of the luminescence produced by a critical reaction of NO with ozone. The measurement of NO_2 is based first on the conversion of NO_2 to NO and then subsequent detection of NO using this well characterized chemiluminescence technique. This conversion is not specific for NO_2 , hence chemiluminescence analyzers are subject to interferences produced by response to other nitrogen containing compounds (e.g., peroxyacetyl nitrate) (PAN) which can be converted to NO . The chemiluminescence technique has been reported to overestimate NO_2 due to these interferences. This is not an issue for compliance, because there are no violations of the NO_2 NAAQS. In addition, the interferences are believed to be relatively small in urban areas.⁹ The air quality trends depicted are in urban locations, and are expected to be reasonable representations of urban NO_2 trends. However, that is not the case in rural and remote areas where air mass aging could foster greater relative levels of PAN and nitric acid and

Figure 2-17. Trend in annual NO_2 mean concentrations, 1988-1997.

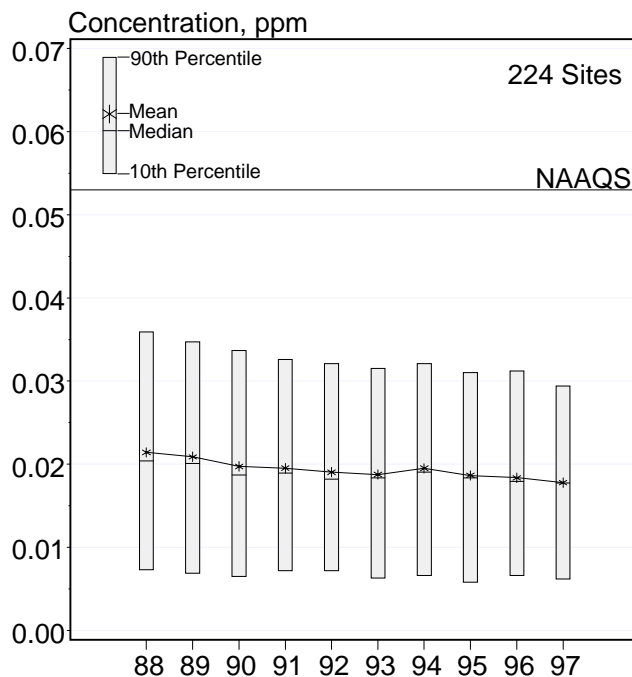


Figure 2-18. Trend in annual mean NO_2 concentrations by type of location, 1988-1997.

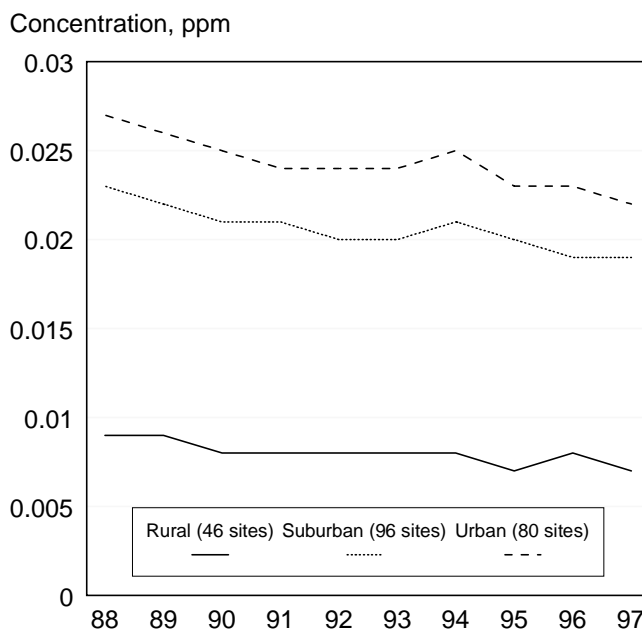
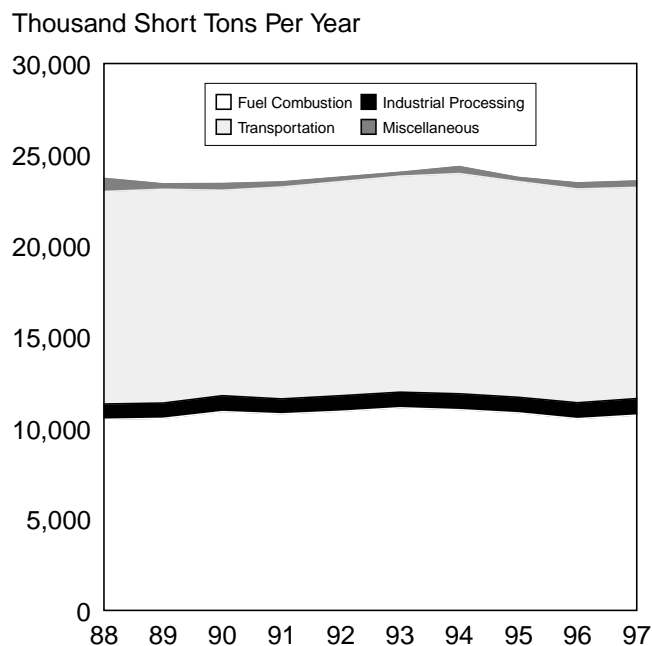
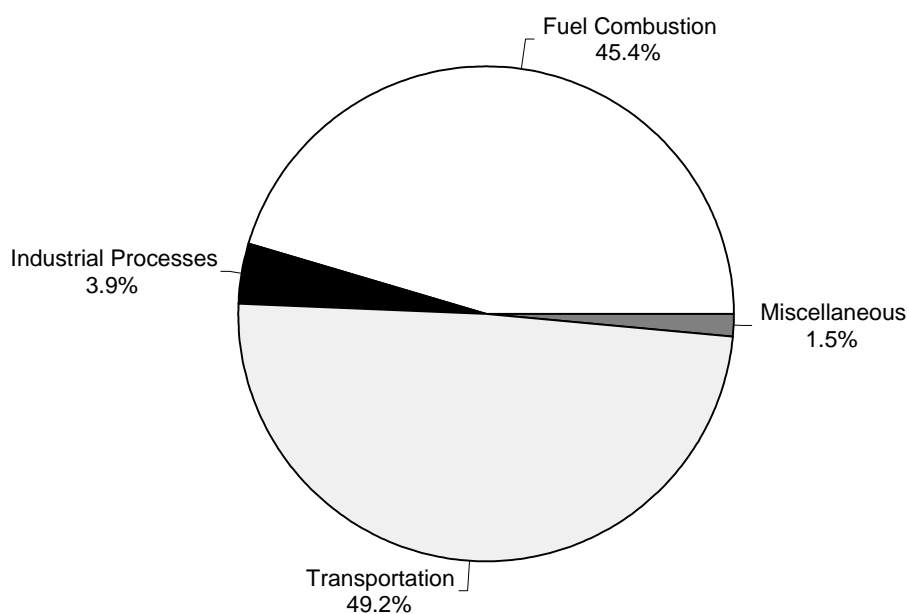


Figure 2-19. Trend in national total NO_x emissions, 1988-1997.**Figure 2-20.** NO_x emissions by source category, 1997.

interfere significantly with the interpretation of NO₂ monitoring data.

Emissions Trends

Figure 2-19 shows the 10-year trend in NO_x emissions. National total NO_x emissions in 1997 are 1 percent lower than the 1988 total, although changes in data availability and methodology between 1989 and 1990 (in the other combustion category) introduce some uncertainty in this comparison.

Emissions from fuel combustion sources are 2 percent higher than the 1988 level, but 4 percent lower than the peak emissions of 1993. Figure 2-20 shows that the two primary sources of NO_x emissions are fuel combustion and transportation. Together these two sources comprise 95 percent of 1997 total NO_x emissions. Because most NO₂ monitors are located in urban, population-oriented areas, that are dominated by mobile sources, the reduction in ambient concentrations (a 14-percent decrease since 1988) more closely tracks the 8-percent decrease in NO_x emissions from highway vehicles. As noted previously in this report, VMT increased 25 percent nationally during the past 10 years. Emissions from coal-fired electric utilities account for roughly one quarter of all NO_x emissions and are not likely to impact most urban NO₂ monitoring sites. Between 1988 and 1997, emissions from these sources decreased 1 percent. Title IV (Acid Deposition Control) of the CAA provides a guideline for NO_x reductions of approximately two million tons from 1980 emissions levels. In 1997, NO_x emissions were reduced 32 percent from 1990 levels at 263 coal-fired units under Phase I of the Acid Rain NO_x Control Program.⁹ These units accounted for

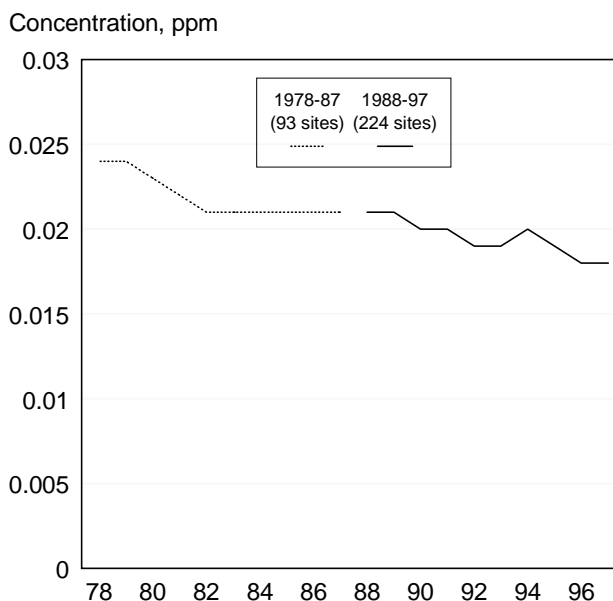
over three percent of total national NO_x emissions in 1997. Between 1996 and 1997, NO_x emissions from these sources increased 1 percent due to greater electrical production.¹⁰ Table A-4 provides a listing of NO_x emissions by major source category.

The significance of the role of nitrogen oxides as a precursor to ozone formation was addressed in a final rule published by EPA in October 1998 (commonly known as the NO_x SIP Call). The rule calls for reductions in summertime NO_x emissions to reduce the regional transport of ozone¹¹ and set (1) a model cap-and-trade program, (2) revised statewide NO_x emission budgets, and (3) proposed revisions to the Acid Rain Program.¹¹ Although the rule does not mandate which sources must reduce emissions, EPA has estimated that reducing NO_x emissions from utilities and large non-utility point sources is one cost-effective strategy available to states. See the ozone section for more information concerning this rule.

National 20-Year Trends

As discussed previously, long-term national ambient air quality trends are difficult to assess because few monitoring sites have operated continuously in the same location for 20 years. Figure 2-21 presents 20-year trends in ambient NO_2 concentrations by combining two separate 10-year trends databases, 1978–1987 (93 sites) and 1988–1997 (224 sites). Nationally, annual mean NO_2 concentrations have decreased in urban areas by approximately 2 percent since 1978. As seen in Figure 2-21, annual mean NO_2 concentrations declined in the early 1980s, were relatively flat during the mid-to-late 1980s, and resumed their decline in the 1990s. The 1997 national composite mean NO_2

Figure 2-21. Long-term trend in annual mean NO_2 concentrations, 1978–1997.



concentration is the lowest level reported during the past 20 years.

Regional Trends

The map in Figure 2-22 shows the trends in NO_2 concentrations during the past 10 years, 1988–1997. The trends statistic is the regional composite mean of the NO_2 annual mean concentrations across all sites with at least 8 years of ambient measurements. Every EPA region (except Region 10 which does not have any NO_2 trend sites) recorded 10-year declines in composite annual mean NO_2 concentrations. Figure 2-22 shows that the largest reductions in composite annual mean NO_2 concentrations occurred in Region 9 (the South Coast of California), followed by Region 1 (the New England states), and the Region 2 states of New York and New Jersey.

1997 Air Quality Status

All monitoring locations across the nation, including Los Angeles, met the NO_2 NAAQS in 1997. This is reflected on the map in Figure 2-23 that displays the highest annual mean NO_2 concentration measured in each county. In July 1998, EPA announced the redesignation of the South Coast Air Basin (the last remaining nonattainment area for NO_2) to attainment for the NO_2 NAAQS.¹²

Data Sources

The NO_2 ambient trends plotting points and emissions totals by source category are listed in Tables A-1 and A-4, respectively. The plotting points for the 20-year trend charts are listed in Table A-9. Table A-11 contains the highest annual mean NO_2 concentration by county in 1997.

Figure 2-22. Trend in NO₂ maximum quarterly mean concentrations by EPA Region, 1988–1997.

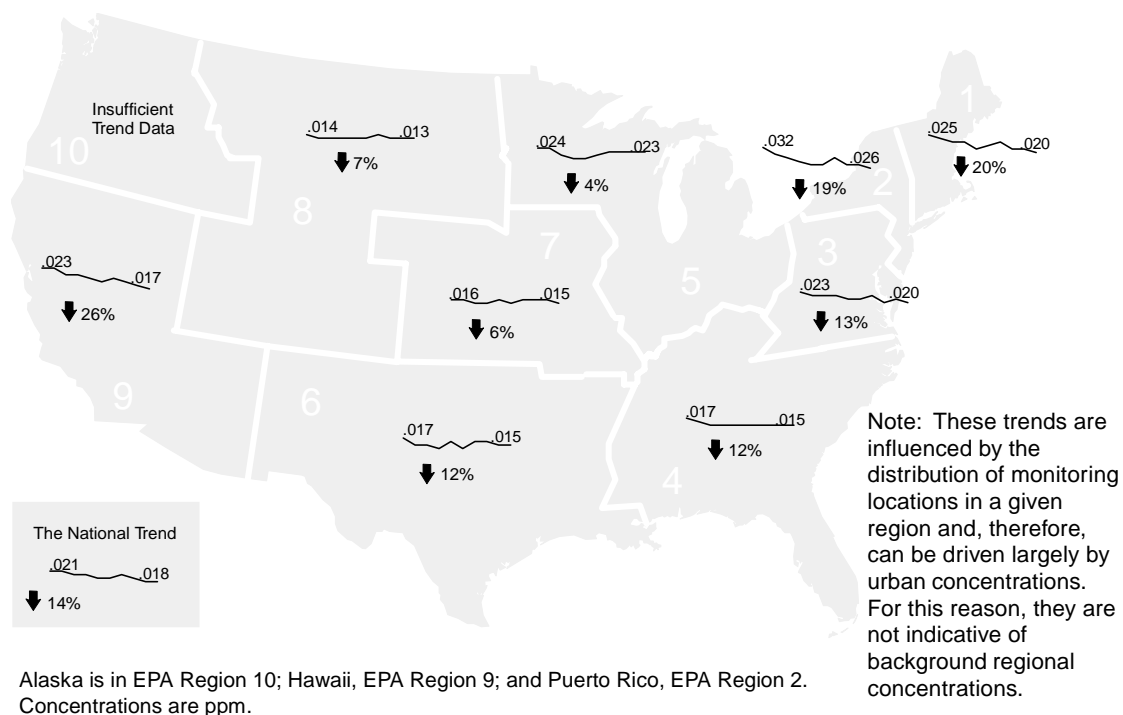
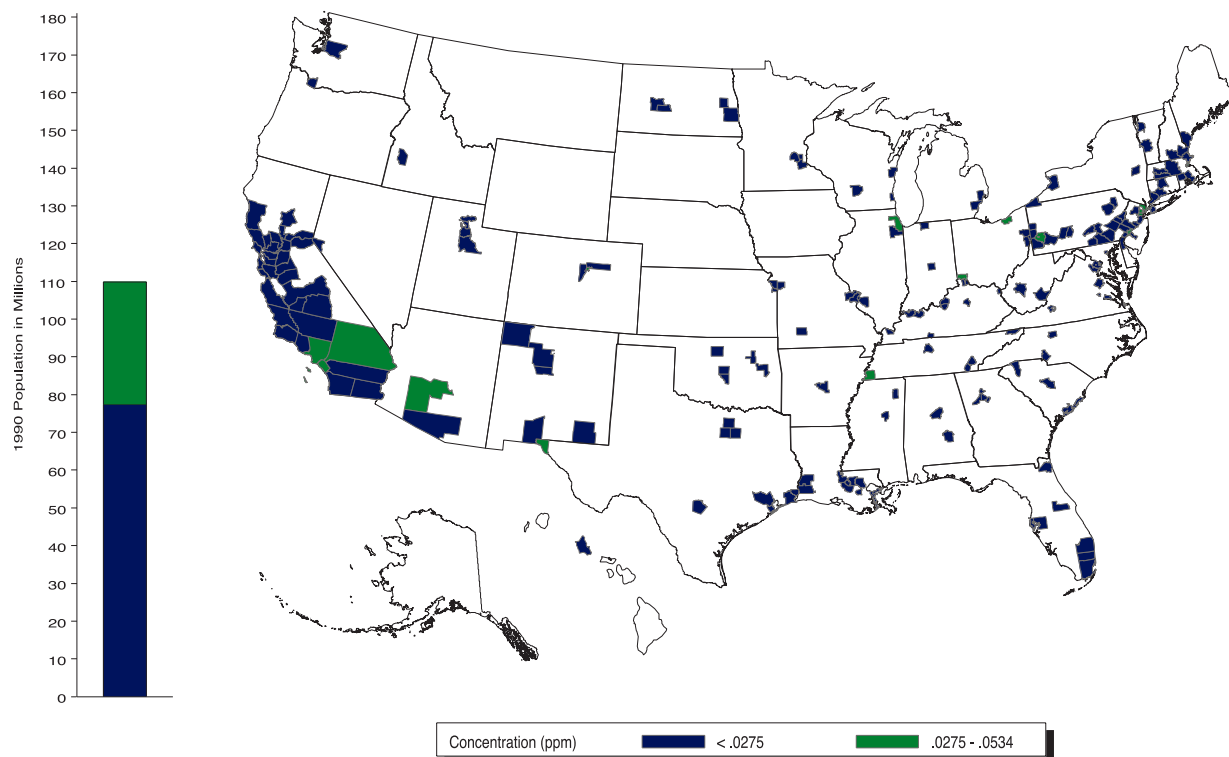


Figure 2-23. Highest NO₂ annual mean concentration by county, 1997.



Ozone

• Air Quality Concentrations

1988–97	19% decrease (1-hr)
	16% decrease (8-hr)
1996–97	no change (1-hr)
	1% decrease (8-hr)

• Emissions

1988–97	20% decrease
1996–97	no change

Nature and Sources

Ground level ozone has remained a pervasive pollution problem throughout the United States. Ozone is formed readily in the atmosphere by the reaction of VOCs and NO_x in the presence of heat and sunlight, which are most abundant in the summer. VOCs are emitted from a variety of sources including: motor vehicles, chemical plants, refineries, factories, consumer and commercial products, other industries, and natural (biogenic) sources. NO_x is emitted from motor vehicles, power plants, other sources of combustion and natural sources including lightning and biological processes in soil. Changing weather patterns contribute to yearly differences in ozone concentrations. Ozone and the precursor pollutants that cause ozone also can be transported into an area from pollution sources found hundreds of miles upwind.

Health and Environmental Effects

Ozone occurs naturally in the stratosphere and provides a protective layer high above the Earth. At ground-level however, it is the prime ingredient of smog. Short-term (1–3 hours) and prolonged (6–8 hours) exposures to ambient ozone concentrations have been linked to a number of health effects of concern. For example, increased hospital admis-

sions and emergency room visits for respiratory causes have been associated with ambient ozone exposures.

Exposures to ozone can make people more susceptible to respiratory infection, result in lung inflammation, and aggravate pre-existing respiratory diseases such as asthma. Other health effects attributed to short-term and prolonged exposures to ozone, generally while individuals are engaged in moderate or heavy exertion, include significant decreases in lung function and increased respiratory symptoms such as chest pain and cough. Children active outdoors during the summer when ozone levels are at their highest are most at risk of experiencing such effects. Other at-risk groups include adults who are active outdoors (e.g., outdoor workers) and individuals with pre-existing respiratory disease such as asthma and chronic obstructive lung disease; within each group there are individuals who are unusually responsive to ozone. In addition, long-term exposures to ozone present the possibility of irreversible changes in the lungs which could lead to premature aging of the lungs and/or chronic respiratory illnesses.

Ozone also affects vegetation and ecosystems, leading to reductions in agricultural and commercial forest yields, reduced growth and survivability of tree seedlings, and increased plant susceptibility to disease, pests, and other environmental stresses (e.g., harsh weather). In long-lived species, these effects may become evident only after several years or even decades, thus, having the potential for long-term effects on forest ecosystems and habitat quality for wildlife and endangered species. Further, ozone injury to the foliage of trees and other plants can decrease

the aesthetic value of ornamental species as well as the natural beauty of our national parks and recreation areas.

Primary and Secondary 1-hour Ozone Standards

In 1979, EPA established 1-hour primary and secondary standards for ozone. The level of the 1-hour primary NAAQS is 0.12 ppm daily maximum 1-hour O₃ concentration that is not to be exceeded more than once per year on average. The secondary standard is identical to the primary standard. To encourage an orderly transition to the revised O₃ standards, the 1-hour standards will no longer apply to an area once EPA determines that the area has air quality data meeting the 1-hour standards. In 1998, EPA revoked the 1-hour O₃ NAAQS in 2918 counties in the United States leaving 225 counties where the 1-hour standard still applies.^{13,14}

Primary and Secondary 8-hour Ozone Standards

On July 18, 1997, EPA established an 8-hour O₃ primary standard to protect against longer exposure periods that are of concern for both human health and welfare (vegetation).¹⁵ The level of the national 8-hour primary and secondary ambient air quality standards for ozone is 0.08 ppm, daily maximum 8-hour average over 3 years. The standards are met when the 3-year average of the annual fourth-highest daily maximum 8-hour ozone concentration is less than or equal to 0.08 ppm.¹⁵ EPA will designate ozone nonattainment areas for the 8-hour ozone NAAQS by July, 2000.¹⁶

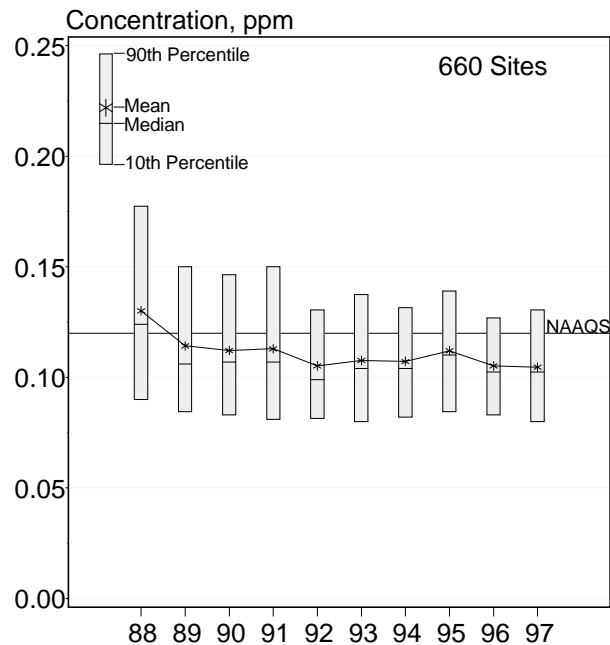
National 10-Year Trends

Because the 1-hour and 8-hour NAAQS have different averaging times and forms, two different statistics are used to track ambient O₃ air quality trends. For the 1-hour O₃ NAAQS, Figure 2-24 presents the national trend in the annual second-highest daily maximum 1-hour O₃ concentration at 660 monitoring sites. The inter-site variability for annual second highest daily maximum 1-hour O₃ concentrations is graphically shown by the 90th percentile, median, composite mean, and 10th percentile concentrations in Figure 2-24. This figure shows that during the past 10-years, higher concentrations have declined more rapidly (the 90th percentile concentration is down 28 percent), while the 1997 national composite average daily maximum 1-hour ozone concentration is 19 percent lower than the 1988 level. The composite mean concentration is unchanged between 1996 and 1997.

Although not shown, the composite average estimated exceedance rate (i.e., the average number of days when the daily maximum 1-hour average concentration exceeds the level of the 1-hour NAAQS) has declined 86 percent since 1988. As noted in previous reports, this statistic, which is simply a count of the number of times the level of the NAAQS has been exceeded, can vary significantly from year to year. Between 1996 and 1997, the national composite mean of the average number of exceedances of the ozone NAAQS declined 30 percent, primarily as a result of the 61 percent decrease in the exceedance rate at sites in California.

For the 8-hour ozone NAAQS, Figure 2-25 presents the trend in the

Figure 2-24. Trend in annual second-highest daily maximum 1-hour O₃ concentrations, 1988-1997.



annual fourth-highest 8-hour daily maximum O₃ concentration at the same 660 sites. The trend in the 8-hour O₃ statistic is similar to the 1-hour trend, although the concentration range is smaller. As measured by the composite mean concentration across all 660 sites, annual fourth-highest 8-hour average concentrations decreased 16 percent since the peak year of 1988. Although, the 8-hour national composite mean concentration decreased 1 percent between 1996 and 1997, the higher concentration sites, as shown by the 90th percentile concentrations, increased 2 percent since 1996.

Ambient O₃ trends are influenced by year-to-year changes in meteorological conditions, population growth, VOC to NO_x ratios, and changes in emissions from ongoing control measures. This 10-year trends period, with peak ozone years at both endpoints, demonstrates the impor-

tance of accounting for year to year variability in meteorological conditions when assessing ozone trends.^{17,18} Previous *Trends Reports* have discussed an EPA statistical model, based on the Weibull probability distribution, that attempts to account for meteorological effects and helps to normalize the resulting trend estimates across years.¹⁸ The model, applied on an individual metropolitan area basis, includes a trend component that adjusts the annual rate of change in ozone for concurrent impacts of meteorological conditions, including surface temperature and wind speed. Figure 2-26 displays the model results for both the 1-hour and 8-hour trends statistics averaged across 41 metropolitan areas. While the ambient monitoring data reflect the year-to-year variability in ozone conducive conditions, the meteorologically adjusted ozone trend provides a better indicator of the impact

of emissions changes. For these 41 metropolitan areas, the adjusted trend for both averaging times shows continued improvement with an average decrease in O_3 concentrations of about 1 percent per year since 1986.

Figure 2-27 shows the 10-year change in ambient ozone concentrations among urban, suburban and rural monitoring sites. The highest ambient O_3 concentrations are typically found at suburban sites, consistent with the downwind transport of emissions from the urban center. During the past 10 years, the composite mean O_3 concentration decreased 23 percent at 117 urban sites and declined by 21 percent at 292 suburban sites. The 1997 composite mean concentration at 234 rural sites is 17 percent lower than the 1988 level.

EPA also announced that it intends to expand the rural ozone monitoring network and to explore opportunities to work with other federal agencies to develop a coordinated and long-term rural monitoring network.¹⁵ One of the ways EPA is accomplishing this is the Clean Air Status and Trends Network (CASTNet) which was developed in response to the CAAA of 1990 requiring implementation of a national network to measure national status and trends. The CASTNet O_3 network, which consists of a total of 69 sites (50 CASTNet and 19 National Park Service (NPS) sites), was designed, in part, to provide information on the distribution of O_3 across rural areas of the United States.

CASTNet sites are considered regionally representative, and thus able to define geographic patterns of rural ozone across the United States. Meteorological variables also are recorded continuously and

Figure 2-25. Trend in annual fourth-highest daily maximum 8-hour O_3 concentrations, 1988-1997.

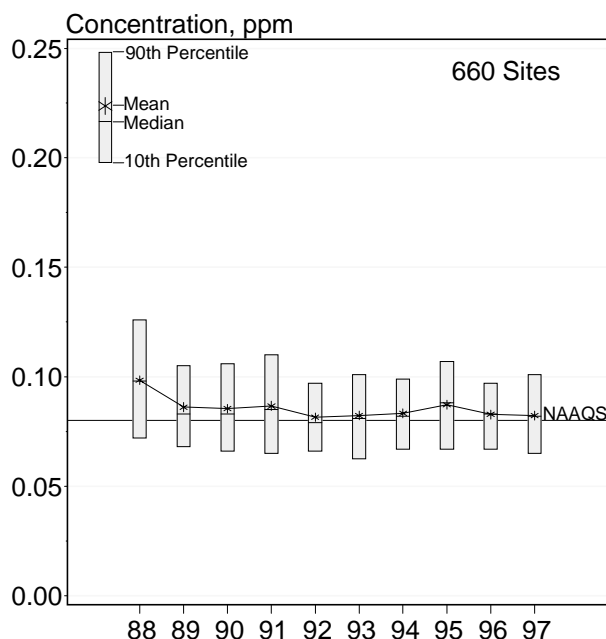


Figure 2-26. Comparison of actual and meteorologically adjusted trends in 1-hour and 8-hour 99th percentile O_3 concentrations, 1988-1997.

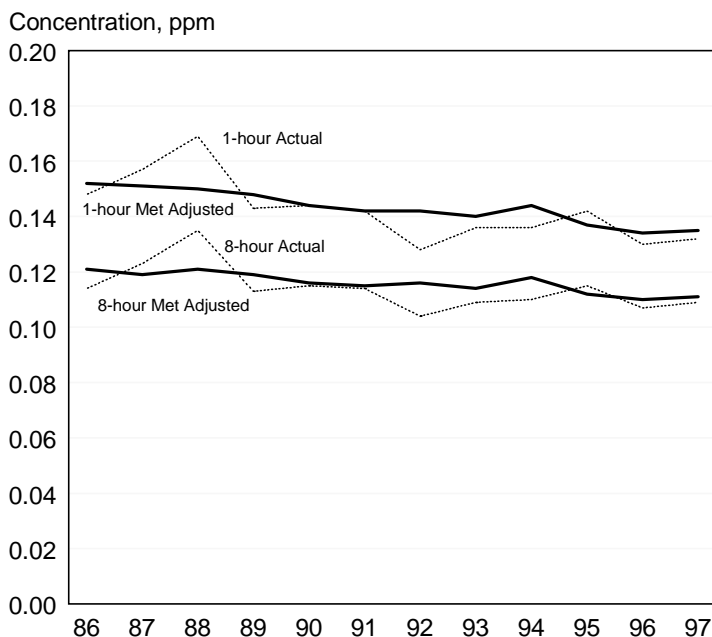
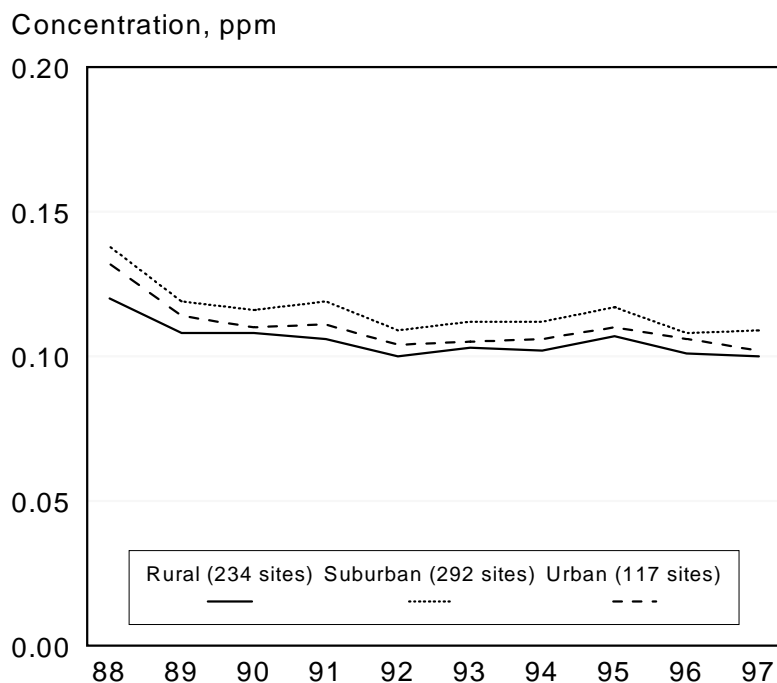
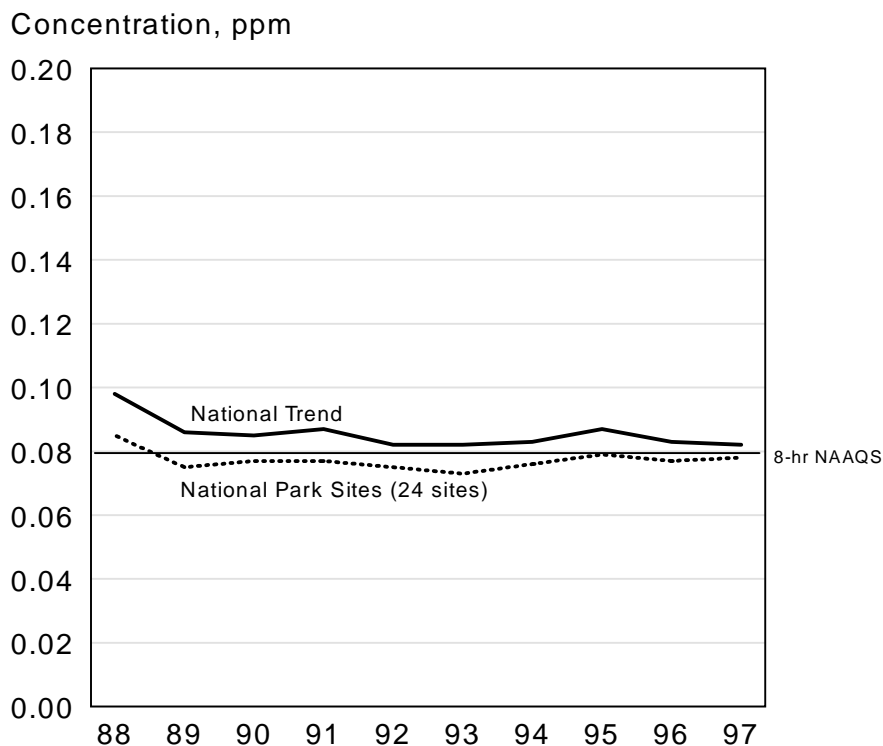


Figure 2-27. Trend in annual second-highest daily maximum 1-hour O₃ concentrations by location, 1988-1997.**Figure 2-28.** Trend in annual fourth-highest daily maximum 8-hour O₃ concentrations in National Parks, 1988-1997.

reported as hourly averages. See Chapter 7: Acid Deposition for more information concerning CASTNet.

Because several other federal agencies have a similar need to understand how ozone impacts the resources they manage, EPA is also working with these agencies to identify better ways to leverage existing monitoring and data collection and analysis efforts. For example, a special subset of rural environments, all national parks and wilderness areas exceeding 5,000 acres, were designated as Class I areas in the 1977 amendments to the CAA. These areas are accorded a higher degree of protection under the CAA provisions for the prevention of significant deterioration. The CAA further directs the federal land managers to protect air-quality related values (AQRVs). Sufficient monitoring data are available to assess 10-year trends in ambient O₃ concentrations at 24 NPS sites. Figure 2-28 compares the 10-year trend in the composite mean of the annual fourth highest 8-hour O₃ concentration at these 24 Class I sites with the national O₃ trend. Non-parametric regression was used to assess the statistical significance of the 10-year trend in 8-hour ozone concentrations for the composite mean across all 24 NPS sites, and at each of the NPS sites. Although the 1997 composite mean O₃ concentration is 8 percent lower than the 1988 value, there is no statistically significant trend in the composite mean O₃ concentration at these NPS sites. On an individual site basis, only two sites, both in the Great Smoky Mountains National Park, had statistically significant upward trends. Although not statistically

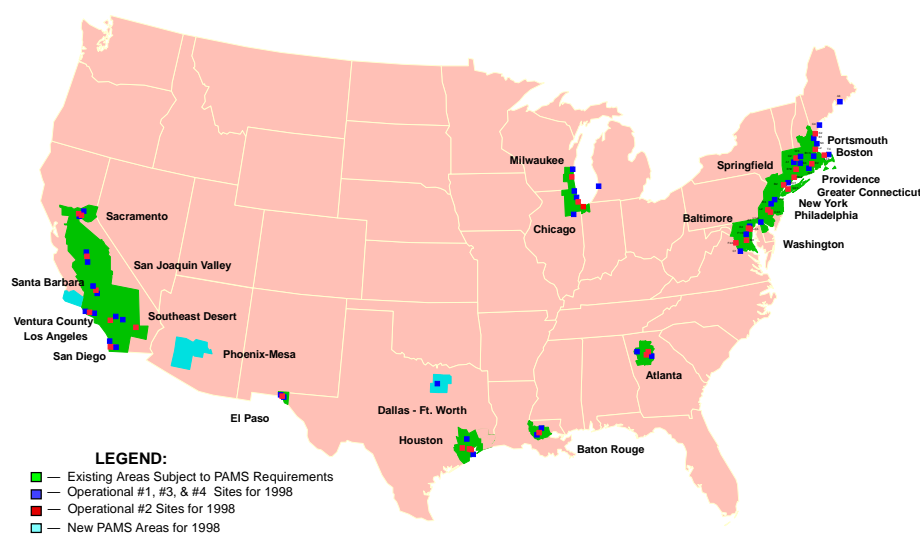
significant, of the remaining 22 sites, 11 sites had downward slopes, 8 upward and 3 sites showed no change. (See Chapter 3: Criteria Pollutants - Metropolitan Area Trends, for a description of the non-parametric regression procedure.)

Enhanced Ozone Monitoring (PAMS)

Section 182(c)(1) of the CAA called for improved monitoring of ozone and its precursors, VOC and NO_x , to obtain more comprehensive and representative data on ozone air pollution. Responding to this requirement, EPA promulgated regulations in February 1993 to initiate the Photochemical Assessment Monitoring Stations (PAMS) program.¹⁹ The PAMS program requires the establishment of an enhanced monitoring network in all ozone nonattainment areas classified as serious, severe, or extreme. Currently, 24 of the remaining 38 nonattainment areas for the 1-hour O_3 NAAQS are subject to PAMS; these areas are identified in Figure 2-29.

Each PAMS network consists of as many as five monitoring stations, depending on the area's population. These stations are carefully located according to meteorology, topography, and relative proximity to emissions sources of VOC and NO_x . Each PAMS network generally consists of four different types of monitoring sites (Types 1, 2, 3, and 4) designed to fulfill unique data collection objectives. Type 1 sites are located upwind of the metropolitan area to measure ozone and precursors being transported into the area. Type 2 sites, referred to as maximum precursor emissions impact sites, are designed to collect data on the type and magnitude of ozone precursor emissions emanating from the metropolitan area and are typically located imme-

Figure 2-29. Metropolitan areas subject to the PAMS program.



diately downwind of the central business district. Type 3 stations are intended to measure maximum ozone concentrations and are sited farther downwind of the urban area than the Type 2 sites. Type 4 PAMS sites are located downwind of the nonattainment area to assess ozone and precursor levels exiting the area and potentially contributing to the ozone problem in other areas. In addition to the surface monitoring sites, each PAMS area also is required to monitor upper air meteorology at one representative site.

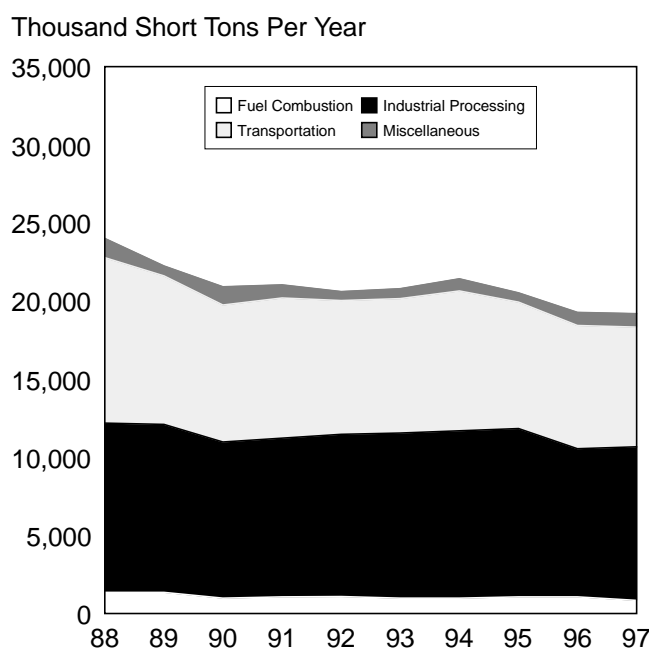
Regulations allow a 5-year transition or phase-in schedule for the program at a rate of at least one station per area per year. The first official year of implementation for PAMS was 1994. As of August 1998, there were 78 operating PAMS sites. The data collected at the PAMS sites include measurements of ozone, NO_x , total non-methane organic compounds (TNMOC), a target list of

VOC species including several carbonyls, plus surface and upper air meteorology. Most PAMS sites measure 56 target hydrocarbons on an hourly or 3-hour basis during the PAMS monitoring season. Included in the monitored VOC species are 10 compounds classified as hazardous air pollutants (HAPs). The PAMS program is the only federally mandated initiative that requires routine monitoring of HAPs; for more information on HAPs see Chapter 5: Air Toxics. All PAMS stations measure ozone, NO_x , and surface meteorological parameters on an hourly basis. In general, the PAMS monitoring season spans the three summer months when weather conditions are most conducive for ozone formation. EPA allows states flexibility in network design and sampling plans in recognition of the fact that each PAMS area has its own unique characteristics and demands. For more information on the PAMS networks, data col-

Table 2-3. Summary of changes in O₃, NO_x and TNMOC at PAMS sites, 1996-1997.

Pollutant	Number of Sites			Median Percent Change
	Total	Up	Down	
O ₃ 2nd daily max 1-hr	69	-	-	1%
NO _x —Summer 6–9am mean	52	8	9	0%
TNMOC—Summer 6–9am mean	42	8	9	-2%

Note: The numbers shown in the “Up” and “Down” categories refer to the number of sites in which the change in summer 6–9am, mean concentrations between the years referenced is a statistically significant increase or decrease (as determined by a t-test with a significance level of .05). The total number of sites (“Total”) may not equal the sum of the corresponding “Up” and “Down” categories.

Figure 2-30. National total VOC emissions trend, 1988–1997.

lected, and analyses of the data, see the EPA PAMS web site at <http://www.epa.gov/oar/oaqps/pams>.

PAMS data provide the opportunity for state and local air pollution control agencies to effectively evaluate ozone nonattainment conditions,

confirm attainment/nonattainment decisions, identify cost-effective control strategies, evaluate population risk exposure, and develop ozone and ozone precursor trends. The measurements have proven extremely valuable in verifying ozone

precursor emissions inventories and in corroborating estimates of area-wide emissions reductions. The data can be used to evaluate, adjust, and provide input to the photochemical grid models used to develop ozone control strategies, as well as demonstrate their success.

Table 2-3 shows second daily maximum 1-hour O₃ concentrations and summer 6–9am mean NO_x and TNMOC concentrations for all reporting PAMS sites for the most recent 2-year period. Morning periods for NO_x and TNMOC are shown since those time frames are generally thought to be an appropriate indicator of anthropogenic emissions. In general, total VOC declined notably between 1994 and 1997, though most of the reductions occurred in the first 2 years, especially between 1994 and 1995. Previous editions of the *Trends Report* highlighted these reductions (as well as corresponding declines in selected VOC species) and attributed them, at least in part, to mobile source controls, specifically the implementation of reformulated gasoline (RFG). Between 1996 and 1997, total VOC only declined slightly (2 percent). NO_x concentrations at PAMS sites were even flatter. The median site concentration is unchanged between 1996 and 1997; only a third of the reporting sites had a significant change in either direction with a fairly even split between sites that showed increases and those that showed declines.

Emissions Trends

Figure 2-30 shows that national total VOC emissions (which contribute to ozone formation) from anthropogenic sources decreased 20 percent between 1988 and 1997. National total NO_x emissions (the other major precursor

to ozone formation) increased 1 percent between 1988 and 1997. Recent control measures to reduce emissions include regulations to lower fuel volatility and to reduce NO_x and VOC emissions from tailpipes.²⁰ The effectiveness of these control measures is reflected in the 28 percent decrease in VOC emissions from transportation sources. VOC emissions from highway vehicles have declined 37 percent since 1988, while highway vehicle NO_x emissions have declined 8 percent since their peak level in 1994. Nationally, the two major sources of VOC emissions are industrial processes (51 percent) and transportation sources (40 percent) as shown in Figure 2-31. Solvent use comprises 66 percent of the industrial process emissions category and 34 percent of total VOC emissions. The emissions totals by source category and year can be found in Table A-5.

As required by the CAA, a cleaner burning fuel (RFG) has been sold since January 1, 1995 in those areas of the country with the worst ozone or smog problems. RFG is formulated to reduce automotive emissions of ozone-forming pollutants and toxic chemicals and is estimated to reduce both VOC and toxic emissions by more than 15 percent.²¹ RFG sold during the summer ozone season has lower volatility than most conventional gasoline.²² The RFG program is mandated year-round in 10 areas of the country (Los Angeles, San Diego, Hartford, New York, Philadelphia, Chicago, Baltimore, Houston, Milwaukee, and Sacramento). Besides these required areas, several other parts of the country exceeding the ozone standard have voluntarily entered the RFG program.²²

Figure 2-31. VOC emissions by source category, 1997.

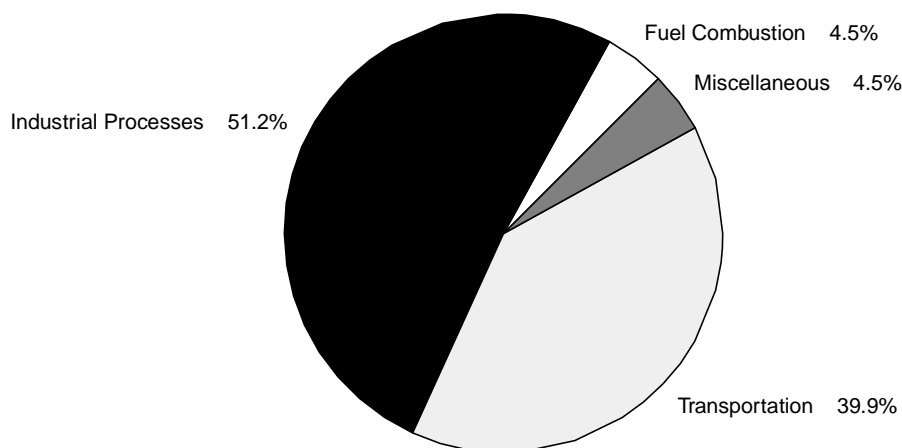


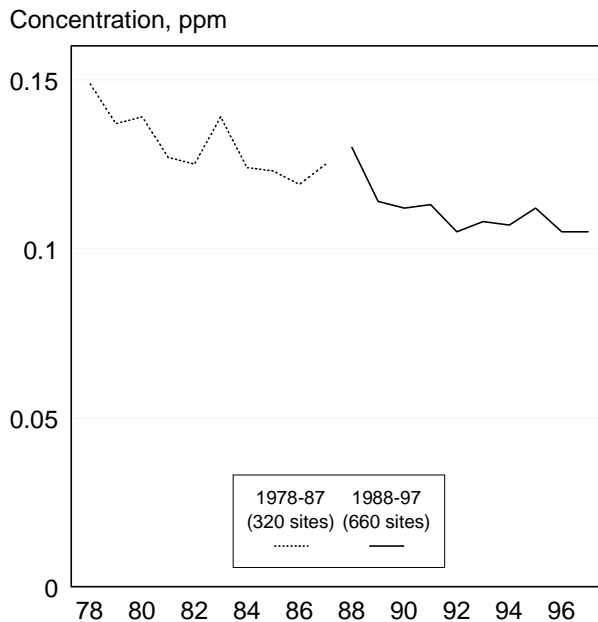
Table 2-4. Biogenic sources of VOC emissions by region.

Region	VOC	Source
Southwestern United States	Isoprene	Oak (mostly), citrus, eucalyptus
	Monoterpenes	Pine, citrus, eucalyptus
Northeastern United States	Isoprene	Oak (mostly), spruce
	Monoterpenes	Maple, hickory, pine, spruce, fir, cottonwood

In addition to anthropogenic sources of VOCs and NO_x, there are natural or biogenic sources of these compounds as well. Table 2-3 shows the different predominant plant species responsible for VOC emissions in different parts of the country for two major biogenic species of concern, isoprene and monoterpenes. Though we are not able to control the level of these natural emissions, when developing ozone control strategies, their presence is an important factor to consider. Biogenic NO_x emissions are

associated with lightning and biological processes in soil.

On a regional basis, biogenic VOC emissions can be greater than anthropogenic VOC emissions. Biogenic NO_x emissions, on the other hand, are less than 10 percent of total NO_x emissions. EPA's estimates of total U.S. VOC emissions from biogenic sources are based on the Biogenic Emissions Inventory System—Version 2 (BEIS2).^{23,24} A recent national estimate for annual total biogenic VOCs from vegetation

Figure 2-32. Trend in annual second-highest daily maximum 1-hour O₃ concentrations, 1978-1997.

is 29 million short tons, while biogenic nitric oxide emissions are estimated at 1.5 million short tons.²⁵ Biogenic emissions are influenced by fluctuations in temperature, with the highest emissions occurring in the summer when temperatures are highest. For example, an increase of 10 degrees Celsius (°C) can result in over a two-fold increase in both VOC and NO_x. Research in the area of biogenic emissions continues, and changes in emission estimates are to be expected and should be viewed with an uncertainty of at least a factor of two.

National 20-Year Trends

Long-term, quantitative ambient ozone trends are difficult to estimate due to changes in network design, siting criteria, spatial coverage and monitoring instrument calibration procedures during the past two decades. For example, in Figure 2-32

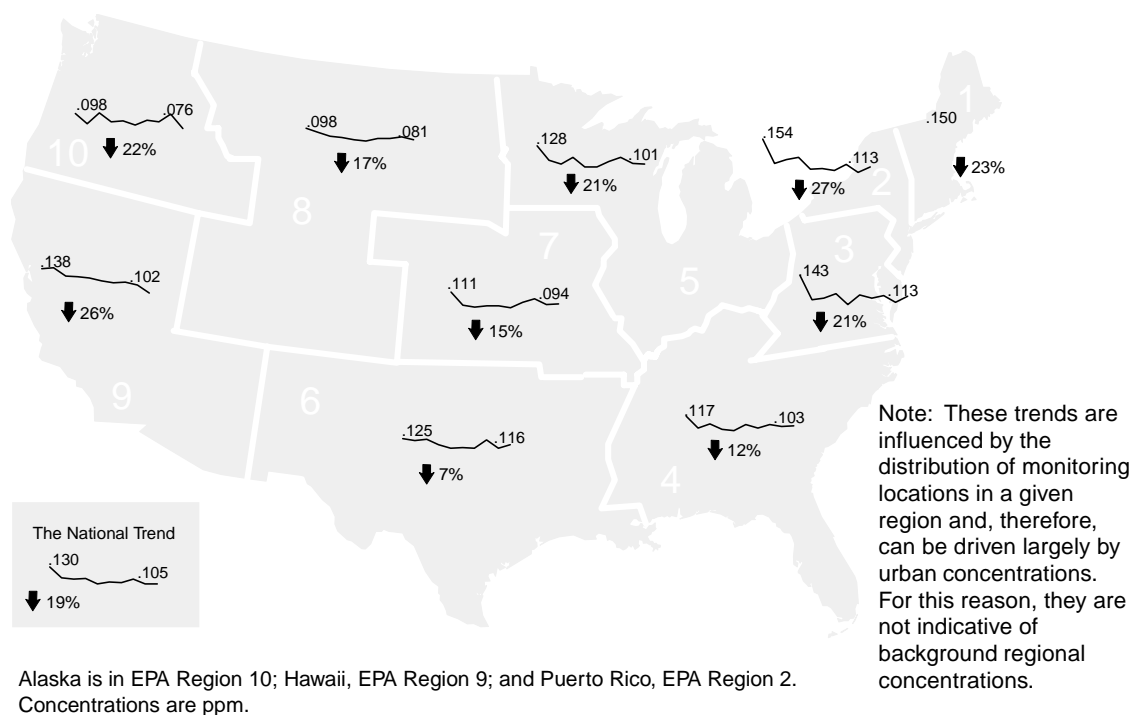
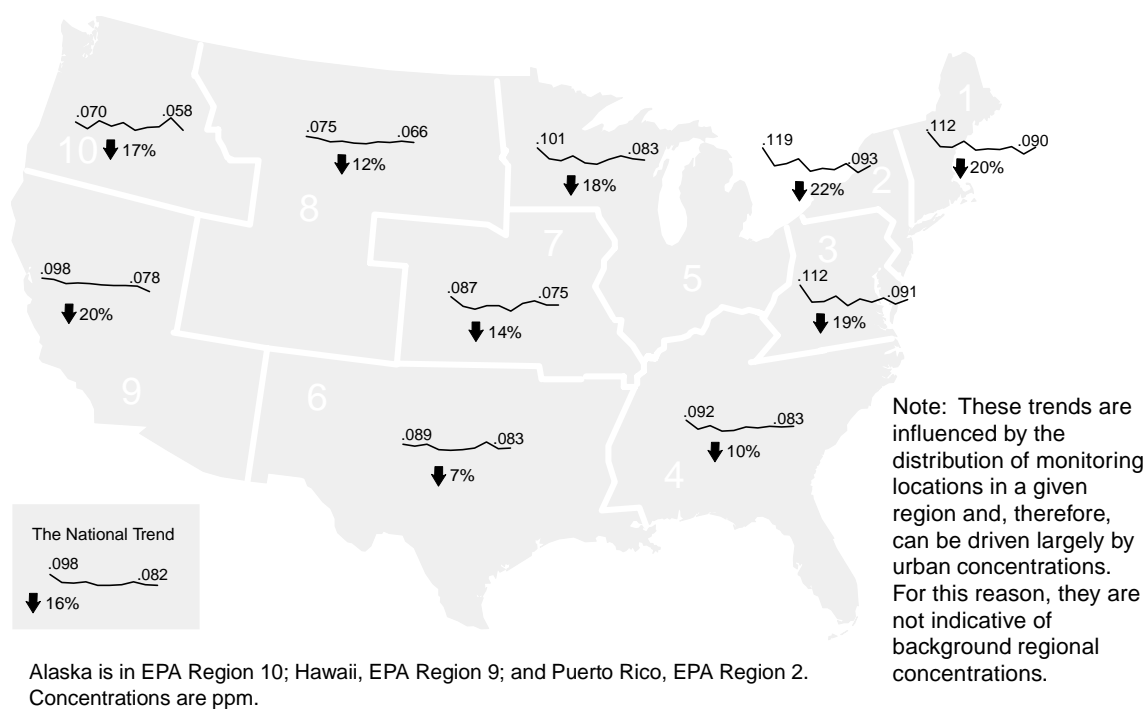
the first year of the early trends period, 1978, corresponds to the use of the old calibration procedure where concentration levels are less certain. Because only a few sites have monitored continuously for two decades, the 20-year trends line in Figure 2-32 is composed of two segments; 238 sites with complete data during the first 10 years, 1978-1987, and 660 sites meeting the data completeness criteria in the most recent 10 years, 1988-1997. Nationally, peak 1-hour O₃ concentrations, as measured by the composite mean of the annual second highest daily maximum 1-hour O₃ concentrations, declined 30 percent since 1978. Figure 2-32 clearly shows the peak ozone years of 1980, 1983, 1988 and 1995.

Regional Trends

The map in Figure 2-32 shows regional trends in 1-hour O₃ concentrations during the past 10 years,

1988-1997. The trends statistic is the composite mean of the annual second-highest daily maximum 1-hour O₃ concentration averaged across all sites in each EPA region with at least eight years of ambient O₃ measurements. Figure 2-34 shows the 10-year trends in the composite mean of the annual fourth-highest daily maximum 8-hour concentration. The trends for both the 1-hour and 8-hour trends statistics are similar, however, the magnitude of the reductions is larger for the annual second-highest 1-hour daily maximum O₃ concentrations as compared to the annual fourth-highest daily maximum 8-hour concentrations. Every EPA region recorded 10-year declines in composite mean 1-hour and 8-hour peak O₃ concentrations.

The greatest improvement in air quality occurred in Northeast, Mid-Atlantic, North Central and Pacific regions. The changes in O₃ concentrations since last year reflect the regional differences in meteorological conditions across the country. Summer 1997 statewide temperature and precipitation ranks are shown in Figure 2-35 based on preliminary meteorological data available from National Oceanic and Atmospheric Administration (NOAA).²⁶ No state was within the top ten warm category and only eight states ranked within the warm third of the temperature distribution. Preliminary data indicate that Summer 1997 was the sixth coolest on record for Georgia and the ninth coolest since 1895 for both Mississippi and South Carolina. Nine states ranked within the top ten dry portion of the historical distribution for Summer 1997 including the fourth driest summer since 1895 for Virginia and Maryland and the fifth driest summer season for

Figure 2-33. Trend in O₃ second maximum 1- hour concentrations by EPA Region, 1988-1997.**Figure 2-34.** Trend in O₃ fourth maximum 8-hour concentration by EPA Region, 1988-1997.

New Jersey. Fifteen other states ranked within the dry-third of the distribution. It was the second wettest summer since records began for California and the ninth wettest summer on record for Montana.²⁶

Addressing The Ozone Transport Issue

In recognition of long-standing regional ozone problems in the Northeastern United States, the 1990 CAAA established the Ozone Transport Commission (OTC) and the Northeast Ozone Transport Region which includes 12 states. Since that time, several other regional groups have formed to study various aspects of the problem and to try to identify acceptable solutions. EPA continues to be a contributor, partner, or interested party in each of these efforts. The most significant recent developments occurred as a result of a 2-year effort known as the Ozone Transport Assessment Group (OTAG), EPA worked in partnership with state and local government agencies in the 37 easternmost states, industry, and academia to address ozone transport. The extensive modeling analysis conducted by OTAG showed the significant contribution of transported precursor emissions to nonattainment of the ozone NAAQS. As a result of OTAG's findings on the role of nitrogen oxides as a precursor to ozone formation, EPA published a rule in October 1998 (commonly known as the NO_x SIP Call) that called for reductions in summertime NO_x emissions to reduce the regional transport of ozone.¹⁰ The NO_x SIP Call sets (1) a model cap-and-trade program, (2) statewide NO_x emission budgets, and (3) proposed revisions to the acid rain program October

1998. More detailed information on the OTAG process and details on information generated by the OTAG workgroups are available on the OTAG web page at <http://www.epa.gov/ttn/otag>.

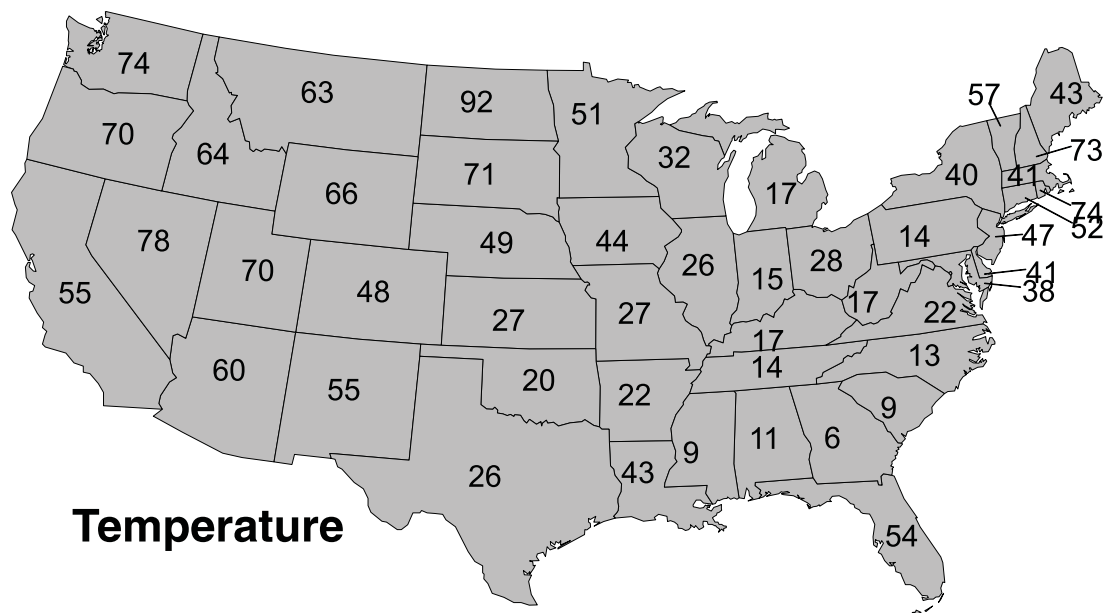
Other regional groups that have addressed regional ozone problems include the Lake Michigan Ozone Study (LMOS), the Southern Oxidant Study (SOS), the Southern Appalachian Mountain Initiative (SAMI), and the North American Research Strategy for Tropospheric Ozone (NARSTO). For more information on these groups, see www.epa.gov/airprog/oar/oaqps/airtrans/regional.html.

1997 Air Quality Status

The map in Figure 2-36 presents the highest second daily maximum 1-hour concentration by county in 1997. The accompanying bar chart to the left of the map reveals that in 1997 approximately 48 million people lived in 77 counties where the annual second daily maximum 1-hour O₃ concentration was above the level of the 1-hour ozone NAAQS. These numbers represent an increase from the totals reported last year (39 million people living in 52 counties) with ozone concentrations above the level of the ozone NAAQS in 1996. As noted previously, meteorological conditions in some regions of the country were more conducive to peak O₃ formation in 1997, than in 1996. The map in Figure 2-36 shows large spatial differences, with higher O₃ concentrations typically found in Southern California, the Gulf Coast, and the Northeast and Northcentral states. Historically, the highest 1-hour concentrations are found in Los Angeles, however, 1997

is the first year that the highest 1-hour concentrations in Houston exceeded the levels recorded in Los Angeles.

Figure 2-35. Summer 1997 statewide temperature ranks (Source: NOAA, 1997).



Note: 1 = coldest/driest ; 103 = warmest/wettest

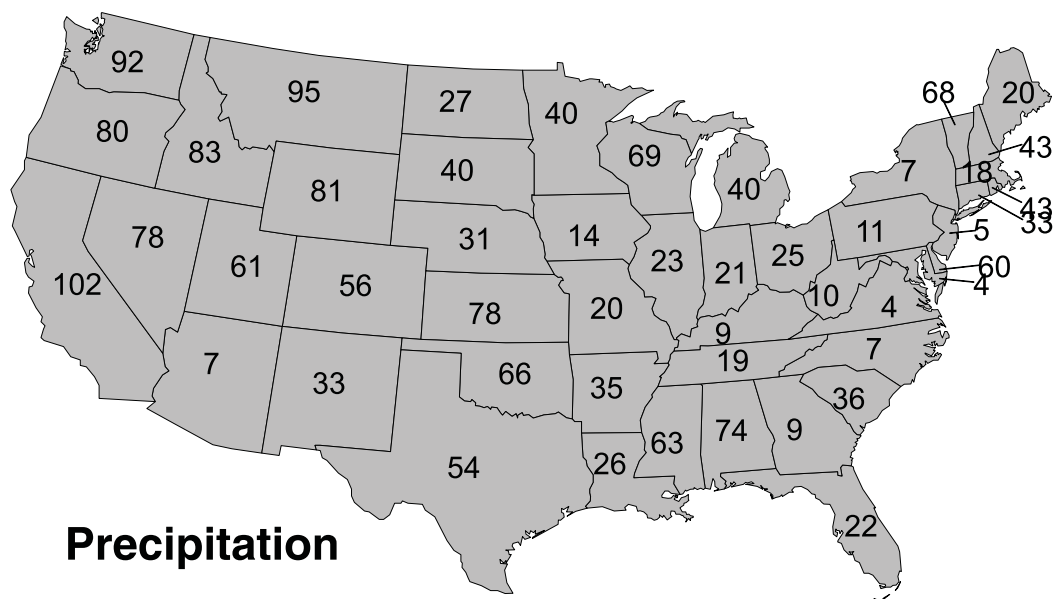
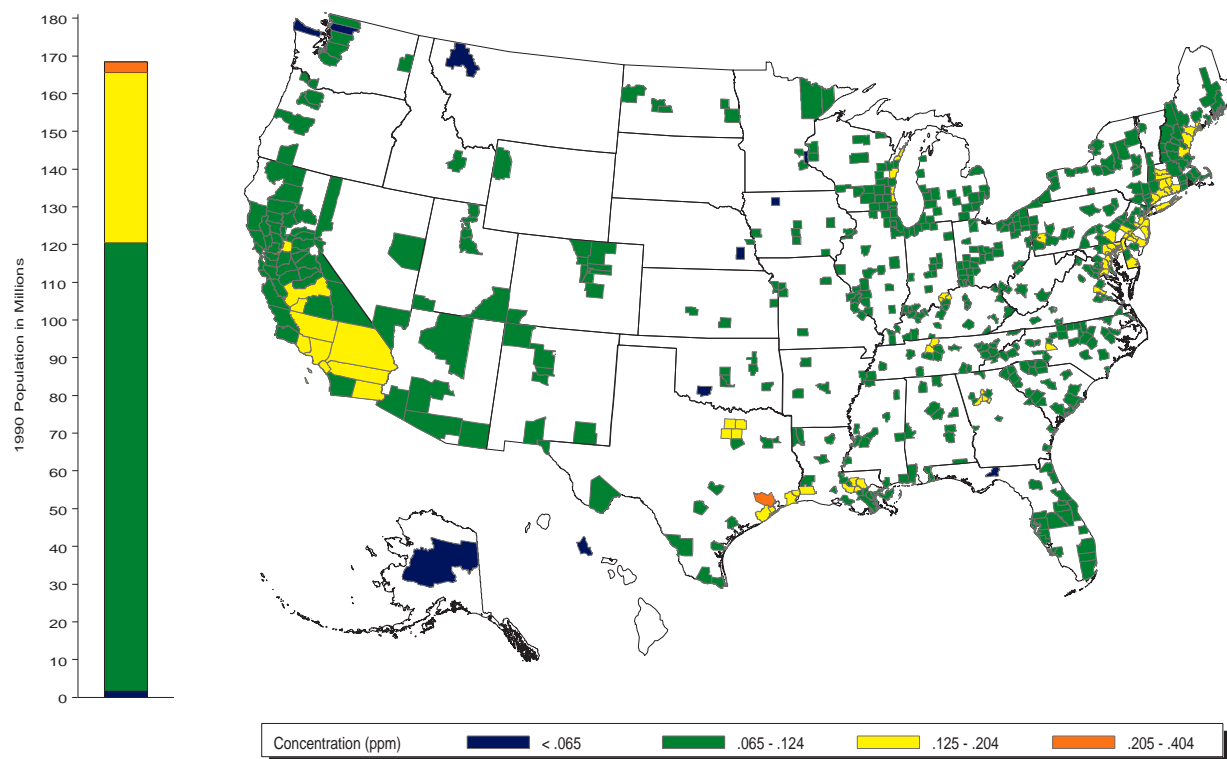


Figure 2-36. Highest second daily maximum 1-hour O₃ concentration by county, 1997.



PARTICULATE MATTER

• Air Quality Concentrations

1988–97	26% decrease
1996–97	1% decrease

• Emissions

1988–97	12% decrease
1996–97	1% decrease

Nature and Sources

PM is the general term used for a mixture of solid particles and liquid droplets found in the air. These particles, which come in a wide range of sizes, originate from many different stationary and mobile sources as well as from natural sources. They may be emitted directly by a source (direct emissions) or formed in the atmosphere by the transformation of gaseous precursor emissions such as SO₂ and NO_x (secondary particles). Their chemical and physical compositions vary depending on location, time of year, and meteorology.

Health and Environmental Effects

Scientific studies show a link between inhalable PM (alone, or combined with other pollutants in the air) and a series of significant health effects. Inhalable PM includes both fine and coarse particles. “Fine” particles are those that are less than 2.5 micrometers in diameter. Those between 2.5 and 10 micrometers are known as “coarse” particles. Both coarse and fine particles can accumulate in the respiratory system and are associated with numerous health effects. Exposure to coarse fraction particles is primarily associated with the aggravation of respiratory conditions such as asthma. Fine particles are most closely associated with such health effects as decreased lung function,

increased hospital admissions and emergency room visits, increased respiratory symptoms and disease, and premature death. Sensitive groups that appear to be at greatest risk to such effects include the elderly, individuals with cardiopulmonary disease including asthma, and children.

In addition, PM causes adverse impacts to the environment. Fine PM is the major cause of reduced visibility in parts of the United States, including many of our National Parks. Other environmental impacts occur when particles deposit onto soils, plants, water, or materials. For example, particles containing nitrogen and sulfur that deposit onto land or water bodies may change the nutrient balance and acidity of those environments so that species composition and buffering capacity change. An ecosystem condition known as “nitrogen saturation,” where additions of nitrogen to soil over time exceed the capacity of the plants and microorganisms to utilize and retain the nitrogen, has already occurred in some areas of the United States.

Particles that are deposited directly onto the leaves of plants can, depending on their chemical composition, corrode leaf surfaces or interfere with plant metabolism. When deposited in sufficient quantities, such as near unpaved roads, tilled fields, or quarries, particles block sunlight from reaching the leaves, stressing or killing the plant. Finally, PM causes soiling and erosion damage to materials, including culturally important objects such as carved monuments and statues.

Primary and Secondary PM₁₀ Standards

The PM trends reported here are based primarily on data collected when the previous NAAQS were in effect. These standards include both short- and long-term PM₁₀ NAAQS. The long-term standard specifies an expected annual arithmetic mean not to exceed 50 µg/m³ averaged over three years. The short-term (24-hour) standard of 150µg/m³ is not to be exceeded more than once per year on average over 3 years. Together, these were the previous primary, or health-based, PM₁₀ standards. The secondary, or welfare-based, standards for PM₁₀ were identical to the primary standards.

The original standards for PM, established in 1971, were for total suspended particulate (TSP) matter. In 1987, EPA replaced the TSP standards with PM₁₀ standards to focus on smaller particles of aerodynamic diameter less than or equal to 10 micrometers. These smaller particles caused the greatest health concern because of their ability to penetrate into sensitive regions of the respiratory tract. The most recent review of the PM standards concluded that still more protection from adverse health effects was needed. In July 1997, the primary (health-based) PM standards were revised to add two new PM_{2.5} standards, set at 15µg/m³ and 65 µg/m³, respectively, for the annual and 24-hour standards, and to change the form of the 24-hour PM₁₀ standard.³⁰ The secondary (welfare-based) standards were revised by making them identical to the primary standards.³⁰ The trends discussion of this section will focus

on the PM_{10} standards that were in place when the majority of the 1988-1997 data presented in this report were collected.

National 10-Year Trends

The first complete year of PM_{10} trends data for most monitors is 1988, so this is the first time that the Trends Report has been able to present a full 10-year air quality trend for PM_{10} . Figure 2-37 shows a 26-percent decrease in the composite average of annual mean PM_{10} concentrations measured at 845 monitoring sites across the country between 1988 and 1997. The downward trend in PM_{10} annual means is apparent, with a leveling off of the trend occurring in the later years. Several factors have played a role in reducing PM_{10} concentrations since 1988. Where appropriate, states required emissions from industrial sources and construction activities to be reduced to meet the PM_{10} standards. Measures were also adopted to reduce street dust emissions, including the use of clean anti-skid materials like washed sand, better control of the amount of material used, and removal of the material from the street as soon as the ice and snow melt. Cleaner burning fuels like natural gas and fuel oil have replaced wood and coal as fuels for residential heating and industrial and electric utility furnaces. The final year change, between 1996 and 1997, shows a decrease of 1 percent. This same general trend can be seen if the sites are grouped as urban, suburban, and rural, as in Figure 2-38. The highest values are generally found at the urban sites, followed closely by the suburban sites. The PM_{10} composite annual mean is significantly lower at the rural sites, which are

Figure 2-37. Trend in annual mean PM_{10} concentrations, 1988-1997.

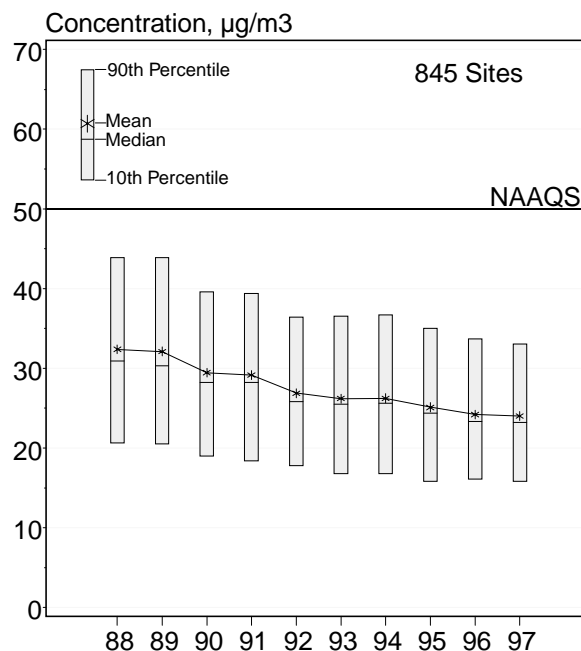
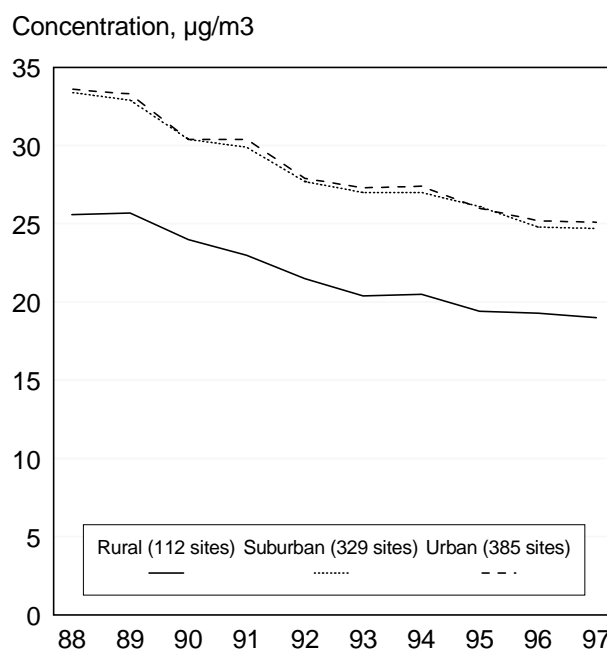


Figure 2-38. PM_{10} annual mean concentration trends by location, 1988-1997.



generally located away from local sources of PM₁₀.

Emissions

Nationally, PM₁₀ direct emissions decreased 12 percent between 1988 and 1997 (see Figure 2-39). Emissions of SO₂, a precursor of PM in the atmosphere, have also been reduced nationally, by 11 percent.

Direct PM₁₀ emissions are generally examined in two separate groups. The first group, shown in Figures 2-39 and 2-40, is the more traditionally inventoried sources. These include fuel combustion, industrial processes, and transportation. Of these, the fuel combustion category saw the largest decrease over the 10-year period (-20 percent), with most of the decline attributable to a decrease in emis-

sions from residential wood burning. Local control programs to curtail the use of residential wood heaters during times when the air was stagnant and to replace old woodstoves with new, cleaner-burning models are responsible for the decrease in residential wood burning, along with lower natural gas and fuel oil prices. Emissions from industrial processes changed very little over the 10-year period, while the transportation category decreased 14 percent. The second group of direct PM₁₀ emissions is a combination of miscellaneous and natural sources including agriculture and forestry, wildfires and managed burning, fugitive dust from paved and unpaved roads, and wind erosion. As Figure 2-41 shows,

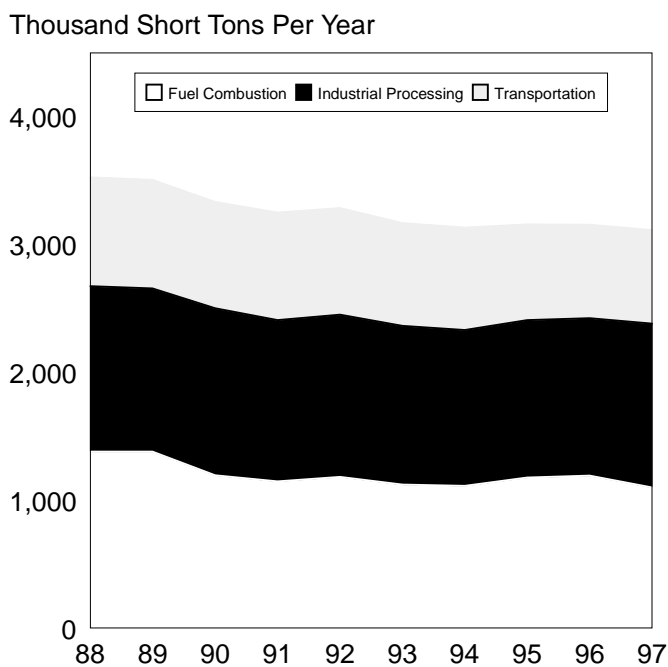
these miscellaneous and natural sources actually account for about 90 percent of the total direct PM₁₀ emissions nationwide, although they can be difficult to quantify compared to the traditionally inventoried sources. Because the emissions in the miscellaneous/natural group tend to fluctuate a great deal from year to year, the trend from one year to the next or over several years may not be particularly meaningful.

Table A-6 lists PM₁₀ emissions estimates for the traditionally inventoried sources for 1988-1997. Miscellaneous and natural source PM₁₀ emissions estimates are provided in Table A-7.

Regional Trends

Figure 2-42 is a map of regional trends for the PM₁₀ annual mean from 1988 to 1997. All ten EPA regions show decreasing trends over the 10-year period, ranging from 19 to 33 percent. The largest decreases are generally seen in the western part of the United States, and the two westernmost regions, IX and X, started at the highest annual mean concentrations back in 1988. In the western states, programs such as those with residential wood heaters and agricultural practices have helped reduce emissions of PM₁₀. Soil moisture levels have also been higher (from more rainfall) in many western states in recent years. In the eastern United States, the Title IV Acid Rain Program has certainly contributed to the decrease in PM₁₀ emissions. The program has reduced SO₂ and NO_x emissions, both precursors of particulate matter in the atmosphere (see the section on SO₂ in this

Figure 2-39. National PM₁₀ emissions trend, 1988-1997 (traditionally inventoried primary PM sources only).



chapter for more on the Acid Rain Program).

1997 Air Quality Status

The map in Figure 2-43 displays the highest second maximum 24-hour PM_{10} concentration by county in 1997. The highest second maximum was recorded in Howell County, Missouri at a monitor adjacent to a charcoal kiln facility. The bar chart which accompanies the national map shows the number of people living in counties within each concentration range. The colors on the map and bar chart correspond to the colors of the concentration ranges displayed in the map legend. In 1997, approximately 5 million people lived in 10 counties where the second highest maximum 24-hour PM_{10} concentration was above the level of the 24-hour PM_{10} NAAQS. When both the current annual and 24-hour standards are considered, there were 8 million people living in 13 counties with PM_{10} concentrations above the PM_{10} NAAQS in 1997.

The Revised Standards

The form of the 24-hour PM_{10} standard changed from the one-expected-exceedance form to a concentration-based 99th percentile form, averaged over 3 years. EPA changed the form of the 24-hour PM_{10} standard from an expected-exceedance form to a concentration-based form because the new form relates more directly to PM concentrations associated with health effects. The concentration-based form also avoids exceedances, regardless of size, from being counted equally in attainment tests. The method for computing the 99th percentile for comparison to the 24-hour standard is found in the

Figure 2-40. PM_{10} emissions from traditionally inventoried source category, 1997.

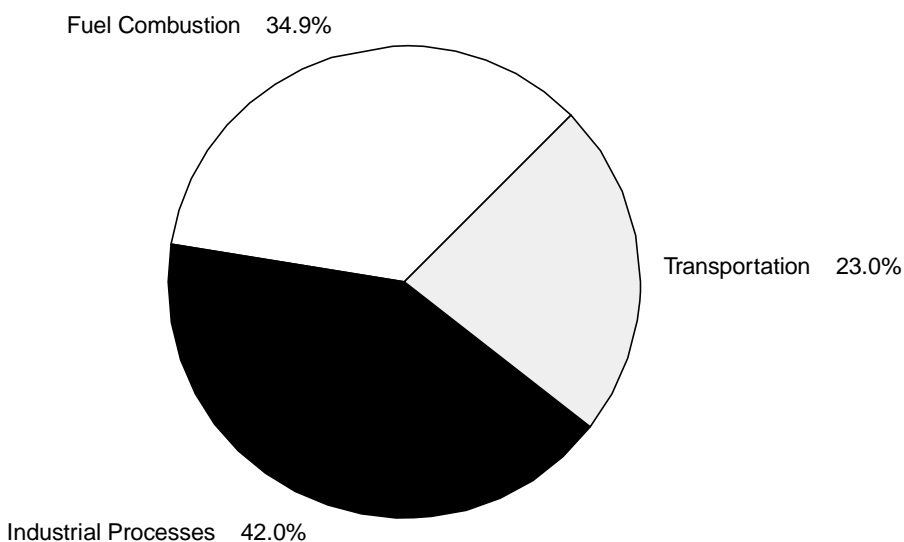


Figure 2-41. Total PM_{10} emissions by source category, 1997.

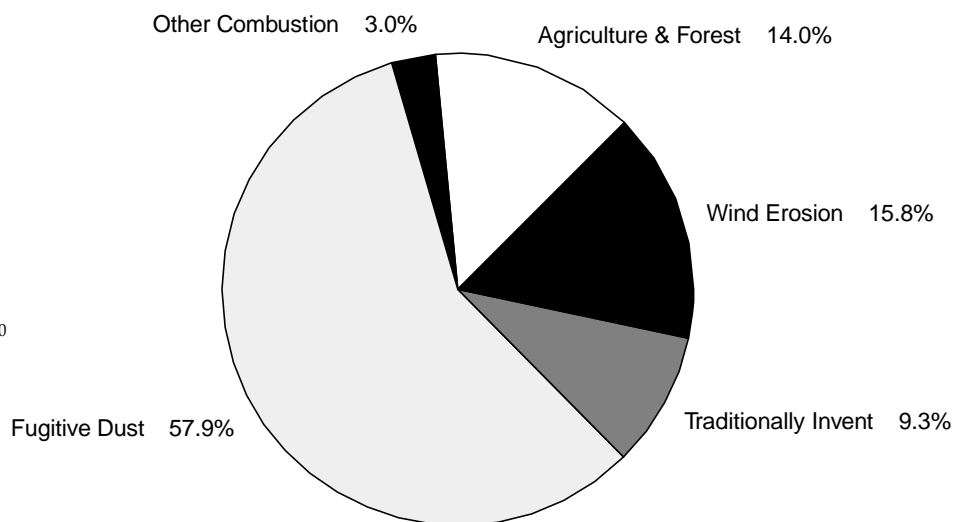
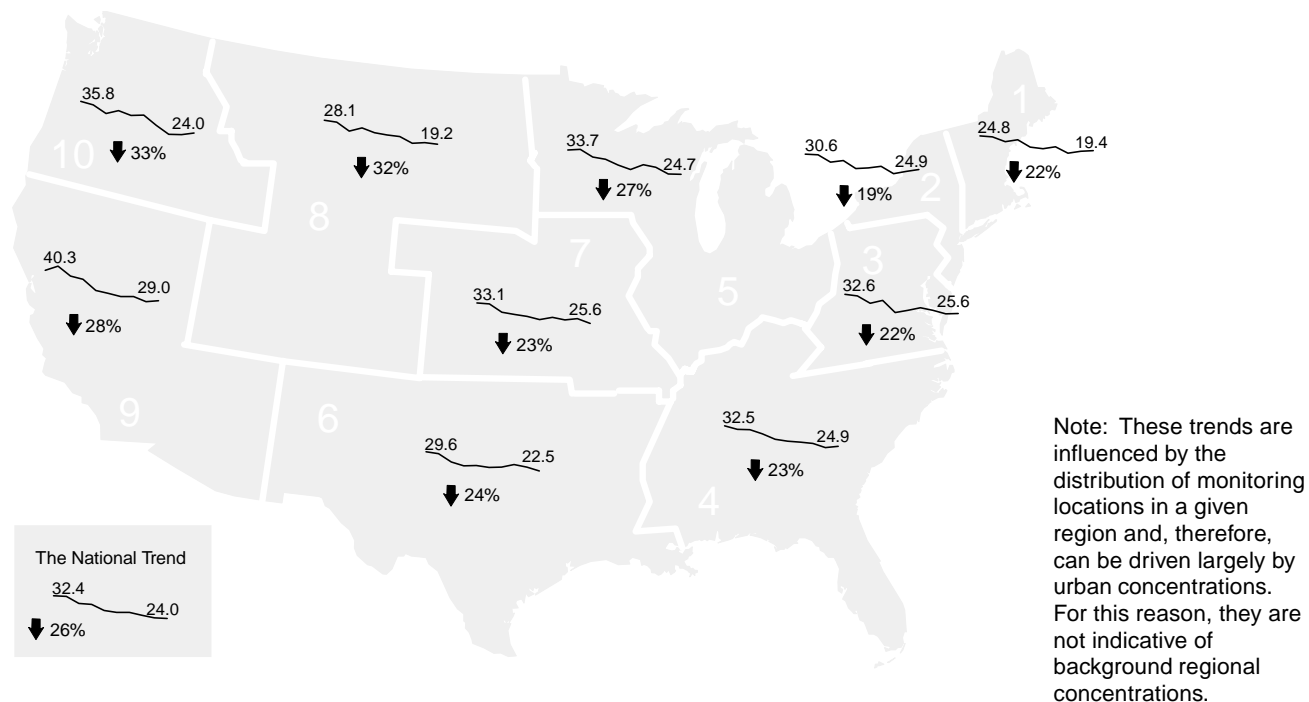
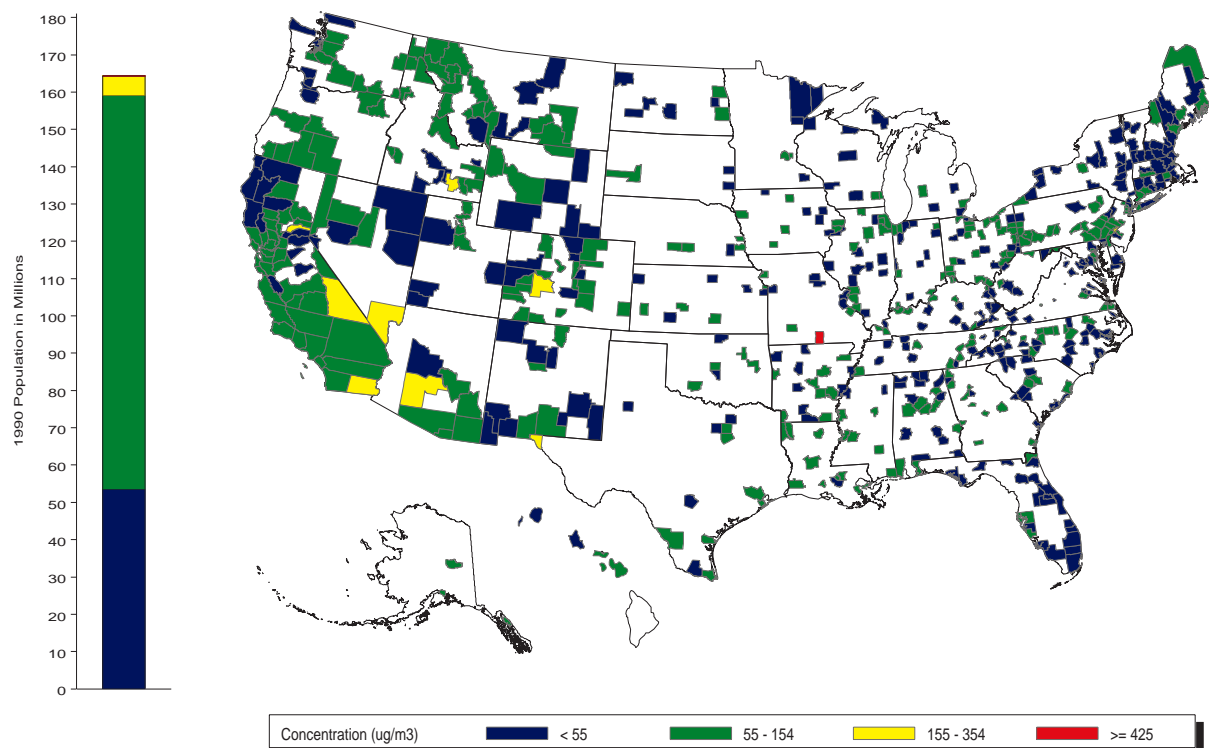


Figure 2-42. Trend in PM₁₀ annual mean concentration by EPA Region, 1988-1997.

Alaska is in EPA Region 10; Hawaii, EPA Region 9; and Puerto Rico, EPA Region 2.
Concentrations are ug/m3.

Figure 2-43. Highest second maximum 24-hour PM₁₀ concentration by county, 1997.

Code of Federal Regulations (40 CFR Part 50, Appendix N).

The form of the 24-hour $PM_{2.5}$ standard is also a percentile form, although it is a 98th percentile. Like PM_{10} , it is averaged over 3 years. The form of the annual standard for $PM_{2.5}$ is a 3-year average of the annual arithmetic mean, just as for the PM_{10} standard. However, unlike PM_{10} , compliance with the $PM_{2.5}$ annual standard may be judged from single or multiple community-oriented monitors reflective of a community-based spatial average. A spatial average can be more representative of community-wide ambient PM exposures.

Beginning in 1998, the revised PM standards require that measurements be reported at conditions of local temperature and pressure (LTP). This is a change from the way PM_{10} data are reported under the pre-existing standards, which specify standard temperature and pressure (STP) for measurement reporting. High altitude or cold regions will see the biggest changes in their concentration levels using LTP, but all monitoring locations will probably see some difference.

Sampling frequencies will change at some locations due to the revised PM standards. New minimum sampling frequency requirements are specified. The number of samples collected in a year has an effect on which concentration value will correspond to the 98th or 99th percentile (i.e. the maximum concentration value collected, the second maximum, and so on). More frequent sampling is desirable because it can force the 98th or 99th percentile to be a value less than the

max, making the statistic more stable.

Characterizing PM_{10} Trends Under the Revised Standards

Figure 2-44 shows a 10-year trend of the average 99th percentile for 845 sites across the country. The 99th percentile shown in the trend is computed by the Aerometric Information Retrieval System (AIRS). The current AIRS uses a slightly different algorithm to compute a 99th percentile than the Code of Federal Regulations (CFR) specifies. The next version of AIRS will correct this inconsistency. Meanwhile, the resulting difference between the two algorithms is insignificant when computing a trend of 845 sites such as Figure 2-43. Any comparisons to the standards for compliance purposes would of course need to use the algorithm specified in the CFR. The trend data show a 25-percent decrease in average 99th percentile concentration between 1988 and 1997.

Characterizing $PM_{2.5}$ Trends Under the Revised Standards

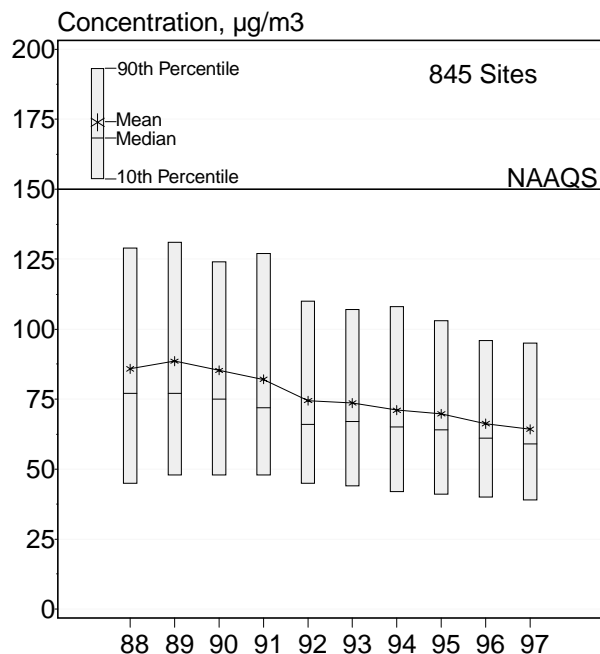
A trend of $PM_{2.5}$ ambient concentration data is not presented here because there are not enough monitors in place at this time to portray an accurate national trend of urban air quality. The network of monitors required for the new $PM_{2.5}$ standard will be phased in over the next few years. For a look at spatial patterns and trends in fine particle concentrations, the reader is directed to the chapter on visibility which documents data derived from the IMPROVE aerosol network. These data are derived from a sampler which is not a Federal Reference Method (FRM) sampler for $PM_{2.5}$. The data

provide a good estimate of urban and nonurban concentrations; however, these data cannot be used for compliance determinations and should be used for preliminary assessments only.

In order to get some idea of the nature of fine PM, some emissions information coupled with ambient data measurements can be examined. EPA is working to improve its $PM_{2.5}$ emission inventory. In the meantime, a general assessment of the emission sources contributing to $PM_{2.5}$ can be obtained by evaluating $PM_{2.5}$ monitoring data. The paragraphs below provide a broad overview of the nationwide concentrations, composition, and sources of $PM_{2.5}$ based on actual $PM_{2.5}$ measurements and the emission inventory of sources contributing within each composition category.

$PM_{2.5}$ is composed of a mixture of particles directly emitted into the air and particles formed in the air from the chemical transformation of gaseous pollutants. The principal types of secondary particles are ammonium sulfate and ammonium nitrate formed in the air from gaseous emissions of SO_2 and NO_x reacting with NH_3 . The main source of SO_2 is combustion of fossil fuels in boilers (including electric utilities), and the main sources of NO_x are combustion of fossil fuel in boilers and mobile sources. Some secondary particles are also formed from volatile organic compounds which are emitted from a wide range of combustion and other sources.

The principal types of directly emitted particles are those that predominantly consist of crustal materials and those consisting of elemental and organic carbonaceous materials resulting from the incom-

Figure 2-44. PM₁₀ trend in the average 99th percentile concentration, 1988-1997.

plete combustion of fossil fuels and biomass materials. The main sources of crustal particles are roads, construction and agriculture. The main sources of combustion-related particles are mobile sources such as diesels, managed burning, open burning, residential wood combustion, and utility, commercial, and industrial boilers.

Figure 2-45 summarizes information from actual measurements of ambient PM_{2.5}. It shows how PM_{2.5} composition varies in both the eastern and western United States. The ambient samples were chemically analyzed to determine the amount of ammonium sulfate and nitrate, crustal material and carbonaceous material. The concentration and composition data are based on at least one year of data from each monitoring location. The

data were collected using a variety of non-federal reference methods and cannot be used to determine compliance with the PM_{2.5} NAAQS.

The figure shows the composition of PM_{2.5} in both urban and nonurban areas of the United States. The composition information represents a range of urban and nonurban locations. The published composition data for the East are somewhat limited, but preliminary information from several recently completed urban studies is included. It shows relatively consistent composition of PM_{2.5} across much of the East. The available information consistently shows that PM_{2.5} in the East is dominated by ammonium sulfate on a regional scale and also by carbonaceous particles emitted directly by combustion processes. Regional concen-

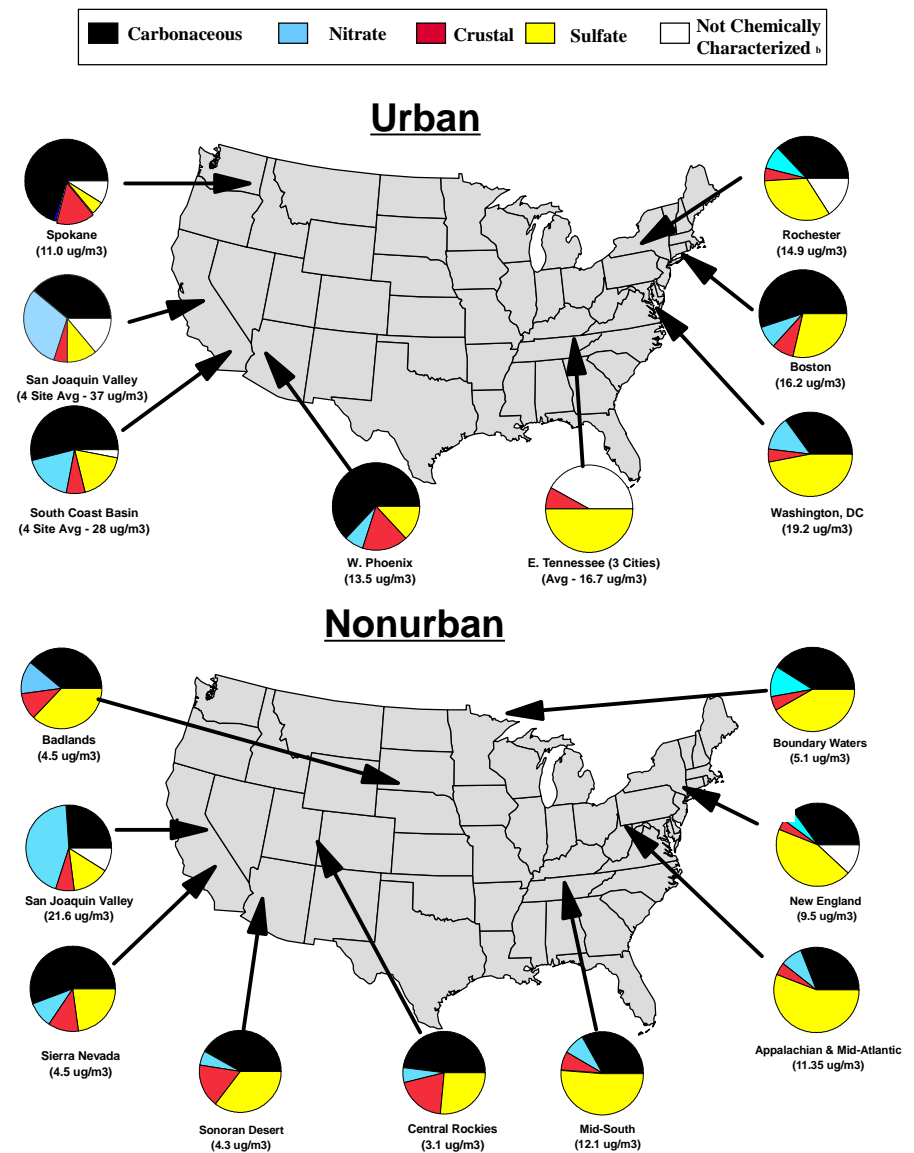
trations of PM_{2.5} are generally higher throughout much of the East, due to the regional influence of ammonium sulfate caused by higher SO₂ emissions throughout much of the East and the ubiquitous nature of combustion processes. (See Chapter 7: Acid Deposition for a description of spatial patterns and trends in sulfate air quality.) The regional concentrations of PM_{2.5} are lower in the western United States than in the East and the composition is more variable. The West differs from the East in two important ways. First, nonurban PM_{2.5} concentrations are much lower in the West than in the East. This is because the East is blanketed regionally by relatively higher concentrations of ammonium sulfate, whereas regional sulfate concentrations in the West are much lower. Second, several western areas, notably the San Joaquin Valley and the Rubidoux area of the South Coast basin have higher ammonium nitrate concentrations. Nitrate concentrations are also higher in nonurban areas of Southern California inland from the South Coast basin. Such pockets of high nitrate concentrations have not been reported in the East. Crustal material is a relatively small constituent of PM_{2.5} in both the West and East, even in arid and agricultural areas such as Phoenix (Arizona) and the San Joaquin Valley of California.

Figure 2-46 depicts the link between sources and the composition components of PM_{2.5}. The EPA has developed a National Emissions Trends (NET) inventory for use in analyzing trends in emissions over time, conducting various in house

analyses for PM, and for use in regional scale modeling.³¹ The NET covers all 50 states and includes point, area, on-road mobile, non-road mobile sources and biogenic/geogenic emissions. Point sources are located individually while county tallies are used for area and mobile source category groups. The inventory includes emissions of SO₂, NO_x, VOC, CO, PM₁₀, elemental carbon and organic carbon. Of these pollutants, only CO is not a contributor to the ambient fine particulate burden. The 1996 NET has been completed for these pollutants and also, a preliminary 1996 NET inventory of PM_{2.5} and NH₃ emissions has been compiled for review by the states. The figure is based in part on information in this preliminary PM_{2.5} inventory and the inventory will be incorporated into the 1996 NET following State review and refinements.

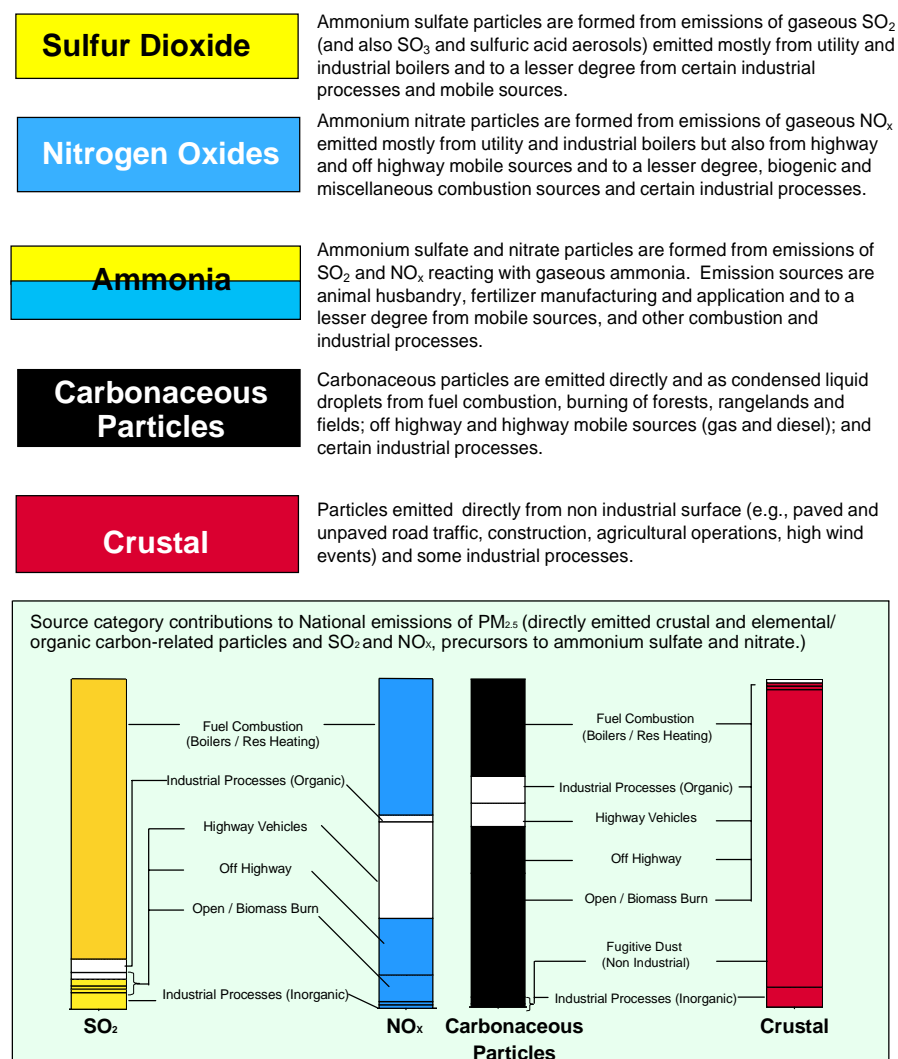
Figure 2-46 provides a link between the sources in the NET inventory and the composition information shown in Figure 2-46. The stacked bar graphs show the relative magnitude of emissions SO₂, NO_x, carbonaceous and crustal-related particles. SO₂ is emitted mostly from the combustion of fossil fuels in boilers operated by electric utilities and industry. Less than 20 percent of SO₂ emissions nationwide are from industrial processes and mobile sources. NO_x emissions are more evenly divided between stationary source and mobile source fuel combustion and biogenic sources are also about 10 percent of NO_x emissions. SO₂ and NO_x also form ammonium sulfate and nitrate in the presence of ammonia under certain atmospheric conditions. Animal husbandry, mobile sources,

Figure 2-45. Summary of information from actual measurements of ambient PM_{2.5}.



a. PM_{2.5} mass concentrations are based on at least one year of monitoring at each location using a variety of non-federal reference methods. They should not be used to determine compliance with the PM_{2.5} NAAQS. Urban pies are based on one site per city or area unless otherwise noted. With exception of the Sierra Nevada and Badlands, nonurban pies represent an average of two or more sites located in the same region.

b. A white segment in a pie indicates that the sum of the constituents (as determined by separate analyses) was less than the gravimetrically determined mass concentration. This could be because study objectives did not require analysis of certain constituents (e.g., no carbon or nitrate analyses for the Tennessee sites) or a variety of technical reasons.

Figure 2-46. PM_{2.5} emission sources.

Note: Composition and source contributions vary among urban areas. Also, some carbonaceous material is formed from organic gases reacting in the atmosphere. The magnitude of these secondary organics is believed small, but more research is needed.

industrial processes and fertilizer application are sources of ammonia. The main sources of carbonaceous (combustion-related) particles are about equally divided among fuel combustion in boilers, biomass combustion and mobile sources. Key biomass sources are wildfires, managed burning and residential

wood burning. Principal mobile sources include both on and off road diesels, gasoline engines and aircraft, railroads and ships. Industrial process emissions will likely be important in some areas as will miscellaneous combustion sources. The main sources of crustal particles are roads, construc-

tion, agriculture and high wind events. Crustal materials are the predominant component of PM₁₀, but Figure 2-46 shows that PM_{2.5} is predominantly comprised of secondary particles and directly emitted carbonaceous particles. The composition (and thus the sources) of PM_{2.5} and PM₁₀ are markedly different because most of the crustal material particles are larger than 2.5 micrometers while almost all of the secondary particles and directly emitted carbonaceous particles are smaller than 2.5 micrometers.

Used together, the figures can give a qualitative feel for the combined influence of specific source types on ambient PM_{2.5} overall (e.g., fuel combustion in boilers, organic and inorganic industrial processes, highway and off highway mobile sources, open burning of waste/biomass and fugitive dust). For example, Figure 2-46 shows that fuel combustion in boilers contributes significantly to both sulfate and carbonaceous mass. Figure 2-45 shows that both sulfate and carbonaceous particles are found in abundance in PM_{2.5} in the East and that carbonaceous particles are also abundant in the West. Thus, preliminary conclusions are (1) that fuel combustion in boilers is a significant contributor to PM_{2.5} in the ambient air and (2) that fugitive dust sources do not appear to play a particularly important role in ambient air samples of PM_{2.5}.

Notes on Data Sources for PM_{2.5}

Composition and concentration data for all non-urban locations were obtained from the Interagency Monitoring of Protected Visual Environments (IMPROVE) except for the New England location which is based on combined non-urban data from IMPROVE and the Northeast States for Coordinated Air Use Management (NESAUM). Washington, D.C. data also were obtained from IMPROVE and the Boston and Rochester data are based on NESAUM. [References: a) IMPROVE, Co-operative Center for Research in the Atmosphere, Colorado State University, Ft. Collins, CO, July 1996. b) Salmon, Lynn, and Glen R. Cass, October, 1997, Progress Report to NESAUM: Determination of Fine Particle Concentration and Chemical Composition in the Northeastern United States, 1995, California Institute of Technology, Pasadena, CA 91125. Draft.] Note that the NESAUM data is still subject to minor revision. The South Coast information is adapted from Christoforou. [Reference: Christoforou, C.S., Lynn G. Salmon, Michael P. Hannigan, Paul A. Soloman and Glen R. Cass, Trends in Fine Particle Concentration and Chemical Composition. Accepted for publication in Journal of Air and Waste Management Association, Pittsburgh, PA.] Phoenix data is from the EPA's Particulate Matter (PM) Research Monitoring Network [Reference: The National Environmental Research Laboratory/ Research Triangle Park PM Research Monitoring Network, U.S. EPA, Research Triangle Park, NC 27711, 1997.] with the exception of the nitrate estimates which were adapted from Desert Research Institute (DRI). [References: (a) PM₁₀ and PM_{2.5} Variations in Time and Space, Desert Research Institute, Reno, NV, October 1995. (b) Watson, John G. and Tom Moore, personal communications with T.G. Pace, December 1997.] The San Joaquin data are from DRI. [Reference: PM₁₀ and PM_{2.5} Variations in Time and Space, Desert Research Institute, Reno, NV, October 1995.] Spokane's composition and concentration data was obtained from Norris. [Reference: 7. Norris, Gary and Jane Koenig, Preliminary Analysis of PM and Daily Emergency Room Visits for Asthma in Spokane, Washington, USA, Presented at International Symposium on Health Effects of PM, Prague, Czech Republic, April 1997.] Eastern Tennessee data was obtained from studies conducted in Knoxville and Chattanooga by the Tennessee Valley Authority and in Nashville by the Harvard School of Public Health.⁸⁹ [References: (a) Tanner, R. (Tennessee Valley Authority) Personal Communication with T.G. Pace, January, 1998. (b) Bahadori, Tina and Helen Suh (Harvard School of Public Health) Personal Communication with T.G. Pace, January, 1998]

Non-urban data are based on averages of several monitoring locations in the region. Urban data are based on only one location in each area and may not represent the entire urban area. The exceptions to this are the South Coast and San Joaquin Valley areas of California where multiple locations are averaged together. In the South Coast basin, Rubidoux recorded the highest average PM_{2.5} and nitrate concentrations. Additional information on the composition of PM_{2.5} within these areas of California is discussed further in Christoforou and DRI. [References: a) Christoforou, C. S., Lynn G. Salmon, Michael P. Hannigan, Paul A. Soloman and Glen R. Cass, Trends in Fine Particle Concentration and Chemical Composition. Accepted for publication in Journal of Air and Waste Management Association, Pittsburgh, PA. and b) PM₁₀ and PM_{2.5} Variations in Time and Space, Desert Research Institute, Reno, NV, October 1995.]

SULFUR DIOXIDE

• Air Quality Concentrations

1988–97	39% decrease
1996–97	4% decrease

• Emissions

1988–97	12% decrease
1996–97	3% increase

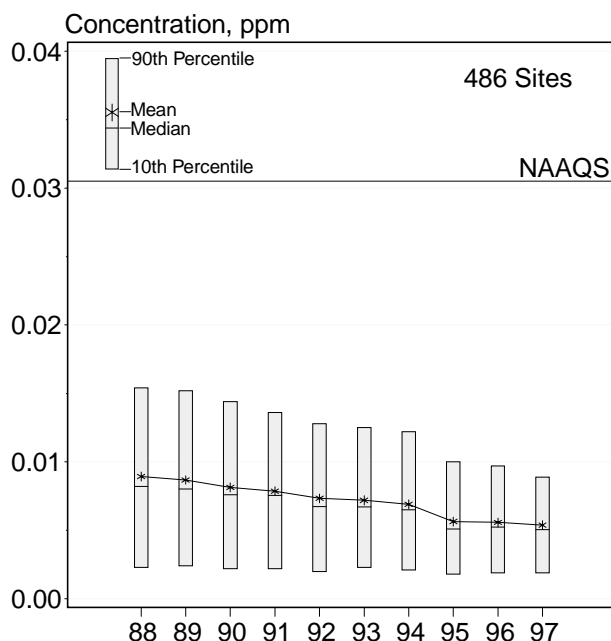
Nature and Sources

SO₂ belongs to the family of SO_x gases. These gases are formed when fuel containing sulfur (mainly coal and oil) is burned, and during metal smelting and other industrial processes. The highest monitored concentrations of SO₂ are recorded in the vicinity of large industrial facilities.

Health and Environmental Effects

High concentrations of SO₂ can result in temporary breathing impairment for asthmatic children and adults who are active outdoors. Short-term exposures of asthmatic individuals to elevated SO₂ levels while at moderate exertion may result in reduced lung function that may be accompanied by symptoms such as wheezing, chest tightness, or shortness of breath. Other effects that have been associated with longer-term exposures to high concentrations of SO₂, in conjunction with high levels of PM, include respiratory illness, alterations in the lungs' defenses, and aggravation of existing cardiovascular disease. The subgroups of the population that may be affected under these conditions include individuals with cardiovascular disease or chronic lung disease, as well as children and the elderly. Additionally, there are a variety of environmental

Figure 2-47. Trend in annual mean SO₂ concentrations, 1988-1997.



concerns associated with high concentrations of SO₂. Because SO₂, along with NO_x, is a major precursor to acidic deposition (acid rain), it contributes to the acidification of soils, lakes and streams and the associated adverse impacts on ecosystems. (See Chapter 7: Acid Deposition). SO₂ exposure to vegetation can increase foliar injury, decrease plant growth and yield, and decrease the number and variety of plant species in a given community. SO₂ also is a major precursor to PM_{2.5}, which is of significant concern to human health (as discussed in the particulate matter section of this chapter), as well as a main pollutant that impairs visibility. (See Chapter 6: Visibility Trends). Finally, SO₂ can accelerate the corrosion of natural and man-made materials (e.g., concrete and limestone) which

are used in buildings and monuments, as well as paper, leather, iron-containing metals and zinc and other protective coatings.

Primary and Secondary Standards

There are both short- and long-term primary NAAQS for SO₂. The short-term (24-hour) standard of 0.14 ppm (365 µg/m³) is not to be exceeded more than once per year. The long-term standard specifies an annual arithmetic mean not to exceed 0.030 ppm (80 µg/m³). The secondary NAAQS (3-hour) of 0.50 ppm (1,300 µg/m³) is not to be exceeded more than once per year.

National 10-Year Trends

The national composite average of SO₂ annual mean concentrations decreased 39 percent between 1988 and 1997 as shown in Figure 2-47,

with the largest single-year reduction (19 percent) occurring between 1994 and 1995.³² The trend has since leveled off, declining only 4 percent from 1996–1997. This same general trend is seen in Figure 2-48 which plots the ambient concentrations grouped by urban, suburban, and rural sites. It shows that the mean concentrations at the urban and suburban sites are consistently higher than those at the rural sites. However, the 1994–1995 reduction in the concentrations at non-rural sites does narrow the gap between the trends. The greater reduction seen in the non-rural sites reflects the fact that the proportion of non-rural sites is greater in the eastern United States, which is where most of the 1994–1995 emissions reductions at electric utilities occurred.³³

Emissions Trends

National SO₂ emissions decreased 12 percent between 1988 and 1997, with a sharp decline between 1994 and 1995, similar to the decline in the ambient concentrations. Unlike the air quality trend, the emissions trend begins to climb again from 1995–1997, as shown in Figure 2-49. This dramatic reduction and subsequent increase is driven by the yearly changes in emissions from the electric utility industry. Much of the increase was caused by units not yet affected by the acid rain program. These units will be in the program, and subject to a national emissions cap, beginning in 2000. The electric utility industry accounts for most of the fuel combustion category in Figure 2-50. In particular, the coal-burning power plants have consistently been the largest contributor to SO₂ emissions as documented in Table A-8 in Appendix A.

Figure 2-48. Annual mean SO₂ concentration by trend location, 1988–1997.

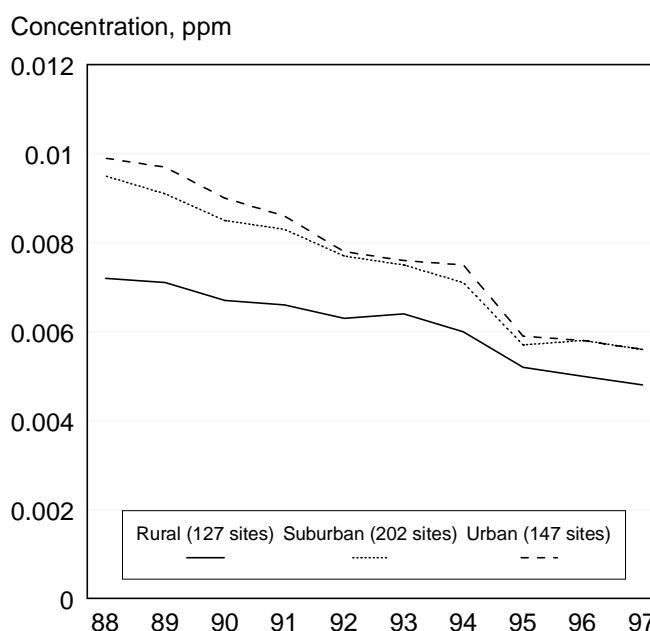


Figure 2-49. National total SO₂ emissions trend, 1988–1997.

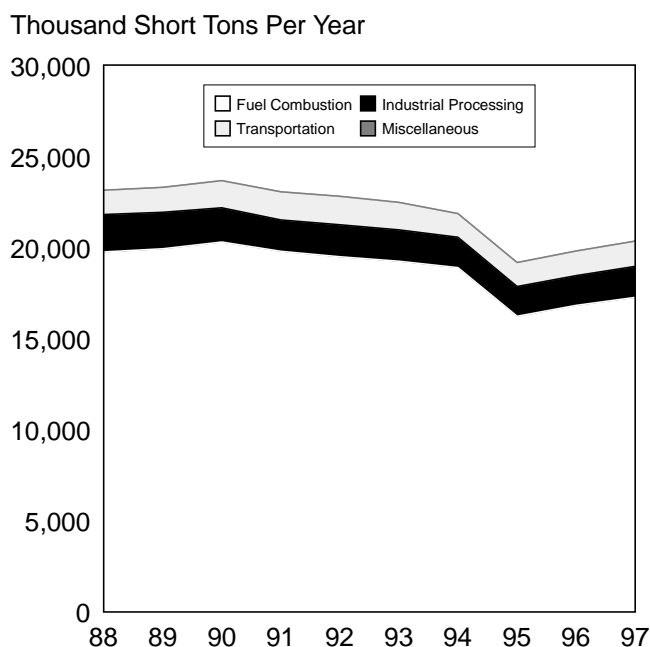
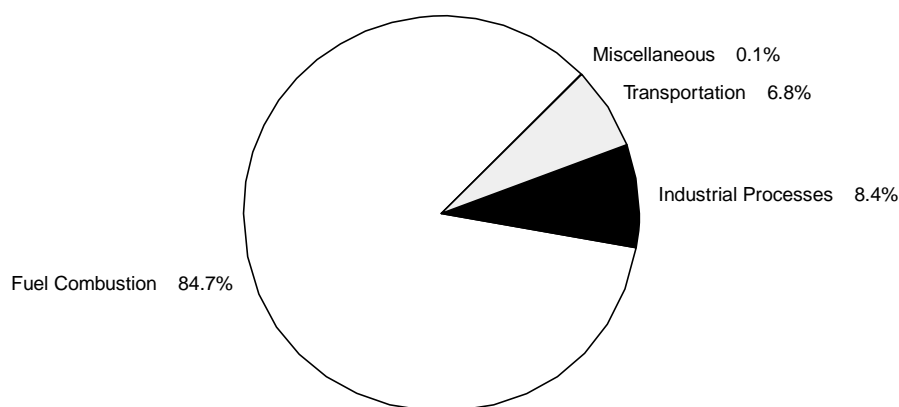
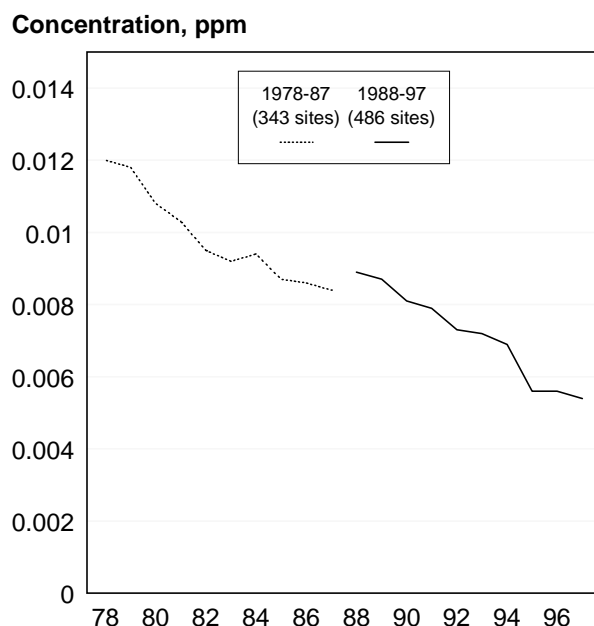


Table 2-5. Total SO₂ Emissions from Phase I units and Non-Phase I units, 1994-97 (thousand short tons).

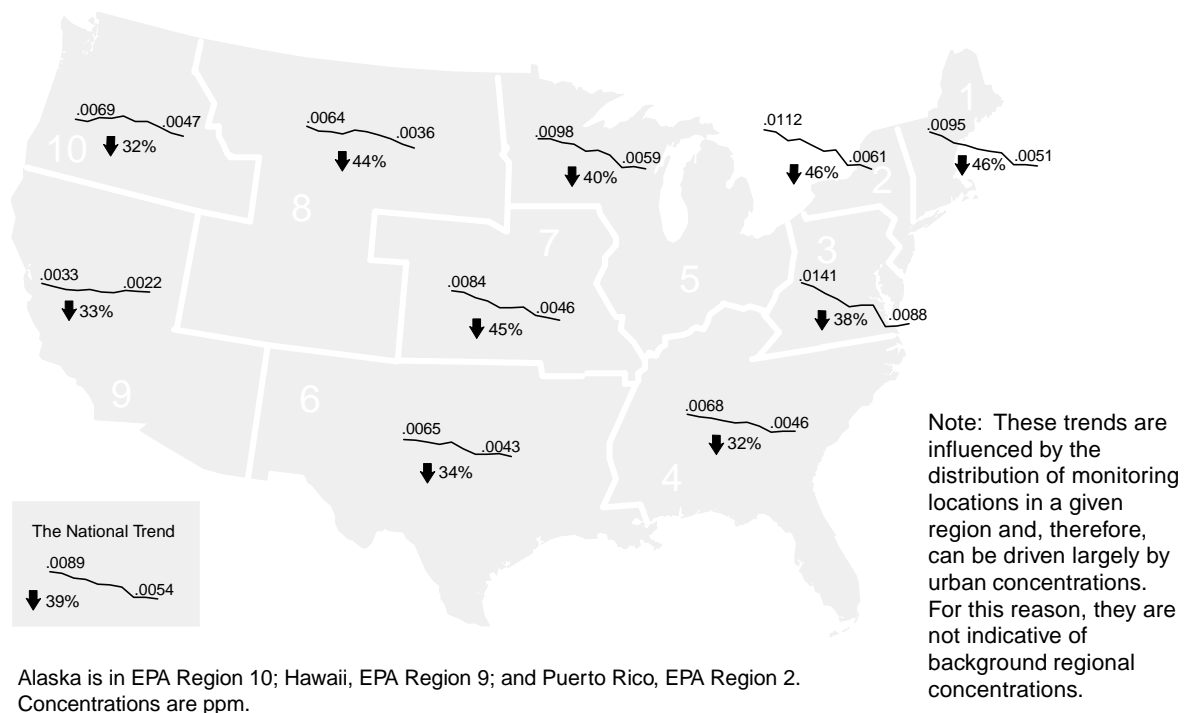
	1994	1995	1996	1997	1994-95	1995-97
Phase I units	6,915	4,938	5,259	5,304	-1,977	+366
Non-Phase I units and Other Units	7,974	7,142	7,373	7,778	-832	+636
All Electric Utility units	14,889	12,080	12,632	13,082	-2,809	+1,002

Figure 2-50. SO₂ emissions by source category, 1997.**Figure 2-51.** Long-term ambient SO₂ trend, 1978-1997.

The Acid Rain Program

The national reductions from 1994-1995 in emissions and ambient concentrations of SO₂ are due mainly to Phase I implementation of the Acid Rain Program. Established by EPA under Title IV of the CAA, the Acid Rain Program's principal goal is to achieve significant reductions in SO₂ and NO_x emissions. Phase I compliance for SO₂ began in 1995 and significantly reduced emissions from the participating utilities.³³ Table 2-5 shows this reduction in terms of Phase I and other units and Non-Phase I and other units.³⁴ The 1994-1995 decrease in total SO₂ emissions from electric utilities is due largely to the Phase I emissions reduction which accounted for 70 percent (1,977 thousand short tons) of the total reduction (2,809 thousand short tons) from electric utilities.

Since 1995, however, total SO₂ emissions from electric utilities have increased. Again, Table 2-4 explains this increase in terms of Phase I units and Non-Phase I units. Most Phase I plants over-complied in 1995 and were able to use their banked emission allowances in 1996 and 1997. As a result, SO₂ emissions have increased slightly at some Phase I sources since the initial reduction in 1995. However, Phase I units account for only 37 percent of the total 1995 to 1997 increase. The majority of the increase is attributed to those units not yet participating in the acid rain program. Most of these units will be included in Phase II of the Program, which begins in 2000. When fully implemented, total SO₂ emissions from electric utilities are capped at 8.9 million tons per year.

Figure 2-52. Trend in SO₂ annual arithmetic mean concentration by EPA Region, 1988-1997.

For more information on the acid rain program, visit <http://www.epa.gov/acidrain>.

National 20-Year Trends

The progress in reducing ambient SO₂ concentrations during the past 20 years is shown in Figure 2-51. While there is a slight disconnect in the trend line between 1987 and 1988 due to the mix of trend sites in each 10-year period, an overall downward trend is evident. In addition to the previously mentioned effects of the Acid Rain Program, these steady reductions over time were accomplished by installing flue-gas control equipment at coal-fired generating plants, reducing emissions from industrial processing facilities such as

smelters and sulfuric acid manufacturing plants, reducing the average sulfur content of fuels burned, and using cleaner fuels in residential and commercial burners.

Regional Trends

The map of regional trends in Figure 2-52 shows that ambient SO₂ concentrations are generally higher in the northeastern United States. The effects of Phase I of the Acid Rain Program are seen most vividly in the northeast. In particular, concentrations fell 21–25 percent between 1994 and 1995 in EPA Regions 1, 2, 3, and 5. These broad regional trends are not surprising since most of the units affected by Phase I of the Acid Rain Program also are located in the north-

east as shown in Figure 2-53. This figure also shows that ambient concentrations have increased slightly between 1995 and 1997 in Regions III and IV where many of the electric utility units not yet affected by the Acid Rain Program are located.

1997 Air Quality Status

The most recent year of ambient data shows that only one area, Buchanan County, Missouri, did not meet the primary SO₂ short-term standard, according to Figure 2-54. The high ambient concentration levels were due to emissions from the local power plant, St. Joe Power and Light Company.

Figure 2-53. Plants affected by Phase I of the Acid Rain Program.

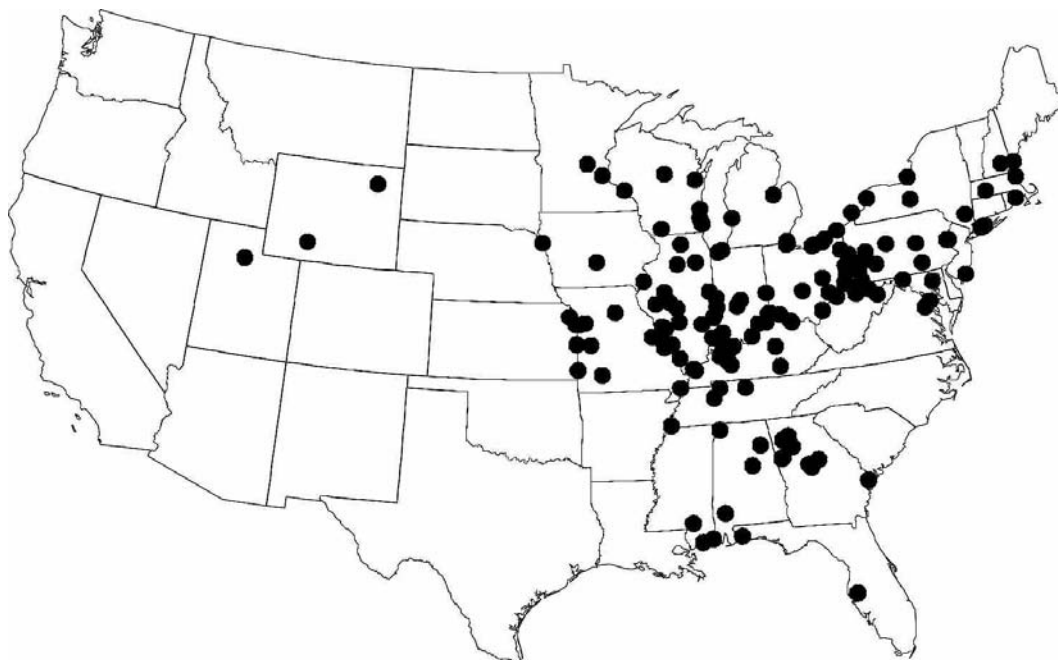
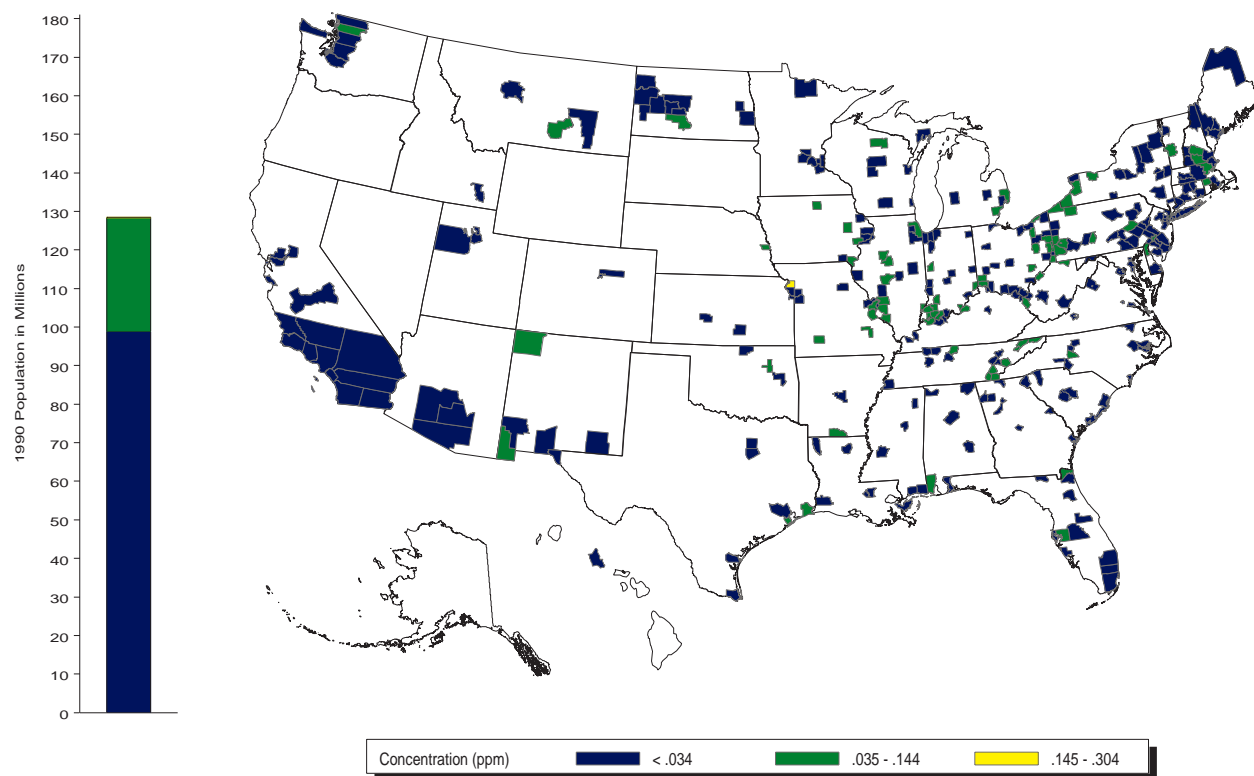


Figure 2-54. Highest second maximum 24-hour SO₂ concentration by county, 1997.



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 10. "1997 Compliance Report," EPA-430-R-98-012, U.S. Environmental Protection Agency, Acid Rain Program, Washington, D.C., August 1998.
- The emission reduction analyses associated with NO_x compliance results in this report focus on 263 of the 265 Phase I NO_x units (representing 170 Table I units and 95 substitution units whose owners chose to participate in Phase I as part of an SO₂ compliance strategy). EPA had determined, as of July 1998, that these units had met the required emission limitation; the two other Phase I units were pending a decision on their alternative emission limitation petitions and determined to be conditionally in compliance.
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 12. "Approval and Promulgation of State Implementation Plans and Redesignation of the South Coast Air Basin in California to Attainment for Nitrogen Dioxide; Direct Final Rule," *Federal Register*, 63 FR 142, Washington, D.C., July 24, 1998.
 13. "Identification of Ozone Areas Attaining the 1-Hour Standard and to Which the 1-Hour Standard Is No Longer Applicable; Final Rule," *Federal Register*, 63 FR 2804, Washington, D.C., June 5, 1998.
 14. "Identification of Additional Ozone Areas Attaining the 1-Hour Standard and to Which the 1-Hour Standard is No Longer Applicable; Final Rule," *Federal Register*, 63 FR 39431, Washington, D.C., July 22, 1998.
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26. "Climate Variations Bulletin: August 1997," *Historical Climatology Series* 4-7, Volume 9, Number 8, National Climatic Data Center, NOAA, Asheville, NC, September 1997.

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28. "Supplemental Notice for the Finding of Significant Contribution and Rulemaking for Certain States in the Ozone Transport Assessment Group Region for Purposes of Reducing Regional Transport of Ozone; Proposed Rule," *Federal Register*, 63 FR 25902, Washington, D.C., May 11, 1998.

29. "Finding of Significant Contribution and Rulemaking for Certain States in the Ozone Transport Assessment Group Region for Purposes of Reducing Regional Transport of Ozone; Final Rule," *Federal Register*, 63 FR 57355, Washington, D.C., October 27, 1998.

30. *National Ambient Air Quality Standards for Particulate Matter: Final Rule*, July 18, 1997. (62 FR 38652), <http://www.epa.gov/ttn/oarpg/rules.html>.

31. *National Air Pollutant Emissions Trends Report*, EPA-454/R-97-011, US EPA, Research Triangle Park, NC 27711, December 1997.

32. The annual mean is used to show trends in national SO₂ air quality because it is a more stable statistic than the 24-hour statistic.

33. *1997 Compliance Report: Acid Rain Program*, EPA-430-R-98-012, U.S. Environmental Protection Agency, Office of Air and Radiation, Washington, D.C., August 1998.

34. The 1995, 1996, and 1997 data for electric utility units in the acid rain program were measured by continuous emissions monitors or equivalent continuous monitoring methods and reported to EPA's Acid Rain Division. The 1994 data are based on fuel use reported to DOE/Energy Information Administration and AP-42 emissions factors. "Phase I units" consist of the 400 units that were subject to Phase I compliance during the first three years (1995-1997) of the acid rain program. These data were obtained from U.S. Environmental Protection Agency, Office of Air and Radiation, Office of Atmospheric Programs, Acid Rain Division, (Mail code: 6204J), Washington, D.C.

